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GEOLOGICAL EVOLUTION OF SOUTH TRANSDANUBIA (HUNGARY) IN PALEOZOIC TIME*

T. SZEDERKÉNYI

SUMMARY

Accepting the opinion about Precambrian age of crystalline basement of South Transdanubia declared by B. JANTSKY [1974], the oldest Paleozoic formations of South Transdanubia can be placed into Ordovician-Silurian period [K. BALOGH, 1974]. Three Ordovician-Silurian rock types are recorded:

1. Strongly folded dark grey siliceous shales with basic tuffs and agglomerates covered by Lower Triassic red sandstones. This rock sequence is interrupted by Carboniferous (?) syenite-porphyric subvolcanic body with uncertain aureole. The unexplored underlying rocks are assumed to pertain to the Precambrian granite mass.
2. The second formation represents a single depositional cycle ranging from coarse grained deposits to limestone with products of ultrabasic and basic volcanism. It forms the oldest Paleozoic part of the so called "Mecsekalja Tectonic Belt" passed along they southern margin of the Mecsek Mountains. These rocks had been superincumbent in Pre-Carboniferous upon the Precambrian "Mórágy Granite Formation" and "Görcsöny Metamorphic Formation". At the beginning of the Variscan Orogeny they had wedged into the underlying Precambrian rocks by a considerable transcurrent faulting, and they suffered a tectonic metamorphism with a slight selective melting. After Pre-Upper Carboniferous denudation of these rocks have taken place the present geological setting.
3. The third formation is formed by ultramafic lava-filled conduits surrounded with Precambrian metamorphites and granites covered by Upper Pannonian and Permian deposits. Rock-material of these vents consists mostly of clinopyroxenites, serpentinites and associated derivatives.

The area affected by powerful movements from the end of the Silurian up to Upper Carboniferous. During this period had been the first manifestation of the Variscan Orogeny caused the wedging of the above mentioned second formation into the "Mecsekalja Tectonic Belt" by transcurrent faulting. It can be placed into Earliest Carboniferous (Bretonian Phase).

During Sudetic Phase a weak granitization had taken place produced microgranite intrusions, aplit veins, K metasomatism as well as presumed syenite-porphyrite near Szalatnak.

At the beginning of Upper Carboniferous the western part of the area formed a basin filled by continental Upper Carboniferous and Permian deposits. The Upper Carboniferous sequence measures more than 1000 m thickness near Téseny—Bogádmindszent covered by Upper Pannonian sediments. It contains some coal beds and coaly siltstone streaks with well determinable flora resembling to the Ruhrland one.

The Upper Carboniferous coal bearing sequence changes gradually into Permian variegated sandstones, siltstones and conglomerates of about 2500 m thickness. The Permian sedimentation was interrupted by an uplift (presumably with local importance) accompanied by a quartzporphyric lava flow attributed to Saalic Phase in Western Mecsek, and by another one in Villány Mts. ranging over the Upper Permian alternating with sandstones.

According to recent views the Upper Carboniferous — Permian sedimentation did not extend more than to so called "Villány—Szalatnak Deep Fracture Zone" in the Eastern direction and line of Drava river in Southern side. The basin of Upper Carboniferous and Permian are made of continental sediments (mainly fluviatile and fewer lacustrine) having a NW—SE general trend.

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At the end of Permian sedimentation a general sinking had taken place which produced a characteristic purplish-red basis conglomerate and sandstone mass due to Pfalz Phase. It is considered as a starting part of the mesozoic sedimentation all over Hungary.

It is important to notice that if the theory of Precambrian age of Görcsöny Metamorphic —, and Mórágy Granite Formations rules out, it will result certain transformation of history of South Transdanubian Old Paleozoic series.

INTRODUCTION

Geological knowledge of ours about Paleozoic of South Transdanubia shows a dissimilar view. The outcrops and not too deeply buried parts of Upper Paleozoic are investigated by boreholes during the uranium prospection and coal mining then a detailed and up to date geological map (scale 1:10.000) of the Mecsek Mts. edited by the Hungarian Geological Survey. Fundamental geological descriptions were recorded by J. BÖCKH [1876] and E. VADÁSZ [1935, 1953, 1960]. From the beginning of the last two decades A. BARABÁS and his coworkers have performed essential work first of all in respect to Permian of South-East Transdanubia.

Apart from several outcrops near Ófalu and Pécs, the Old Paleozoic rocks are covered by younger formations and they are studied mainly by deep drillings. Contrasting with Late Paleozoic, these rocks — according to present-day knowledge — do not contain ore minerals in remarkable quantity. Exploration of them could be regarded as random-like events (geophysical controll drillings and several oil prospecting wells drilled into several Upper Pannonian anticlines) giving isolated and uncertain data. So the geological examination, correlation and exact interpretation being in an initial stage all over the Transdanubia. There is only a comprehensive interpretation of Old Paleozoic in relation with Precambrian crystalline basement on one hand [B. JANTSKY, 1974] and stratigraphic, petrographic and geochemical evaluation [T. SZEDERKÉNYI, 1962, 1970, 1974] as a part of monographic elaboration, on the other hand.

Stratigraphy and chronology of Paleozoic formations were determined on the basis of different geological considerations without acceptable fossils. The stratigraphy is mostly lithostratigraphy with a fairly good accuracy in the South–East Transdanubian Paleozoic. The situation in this respect is much more unmaintainable in the Paleozoic of the area between Balaton lake and Mecsek Mts. Apart from few exceptions, the age of the groundfloor in this area — explored several boreholes — sometimes variously modified. E.g. Igal limestone affected a powerful alteration because of Helvetian and Tortonian volcanism (in the neighbourhood) it was in the first time described as Old Paleozoic, then changed into Carboniferous, lastly it is regarded as an Anisian limestone. Or limestone Karád–1 deep drilling, where — according to original documents — the Upper Carboniferous age was detected only in fragments of Miocene breccia. In spite of this fact the Hungarian geological literature is full of references to this Upper Carboniferous, moreover this datum is the cardinal point some paleogeographic and tectonic ideas referring to the Upper Carboniferous era.

It is an unfortunate fact that fundamental establishments, research conceptions, important paleogeographic conclusions as well as industrial investigations based on defective or uncertain data were born. After some unsuccessfulnesses it has arisen the necessity of a general revision connected with Old Paleozoic of South Transdanubia, mainly on demand of the petroleum and ore mining. This revision-work is in progress. Consequently this paper throws some lights on the geological evolution of the region in accordances with the new developed results and news.

Many of the data collected are not yet available in published form, being either uranium and oil company files and other places or awaiting publication, so that some of the opinions expressed here, may be modified in future.

GROUNDFLOOR OF PALEOZOIC OF SOUTH TRANSDANUBIA

(A review)

Crystalline rocks give the bedrock for Paleozoic or younger formations of South Transdanubia. They can be divided into two parts: metamorphites and granitoids. The latter occupies an elongated zone within metamorphites striking Northeast, from Szigetvár up to the centre of Great Hungarian Plain. This trend is generally characteristic in the Pannonian Mass except the metamorphic basement of Drava Basin where Northwest trend prevailes without granitoids [T. SZEDERKÉNYI, 1974]. The metamorphism is dated on Early Proterozoic due to the effect of Gottidic Orogeny and the age of granitisation assumed to be Late Proterozoic by B. JANTSKY [1974]. Without counterproof we accept this idea and found our Old Paleozoic classification on this statement.

EVENTS OF CALEDONIAN TECTONIC UNIT

There is no proof to the existence of Cambrian and Ordovician rocks on South Transdanubia. At present the oldest known Paleozoic formation is located Northeast Mecsek Mts. near Szalatnak, aged Silurian based on several badly preserved fossils [J. ORAVECZ, 1964]. It is possible that the unexplored introductory part of this group can be placed to Ordovician [K. BALOGH, 1974].

1. *Szalatnak Siliceous Shale Formation*

It is extended to about 300 km² area in Northeast Mecsek Mts. between Szekszárd and Dombóvár *(Fig. 1)* covered by Lower Triassic, Miocene or Upper Pannonian deposits. In the centre of this area the Formation was explored by six boreholes which produced a strongly folded sequence with tripartition which can be studied best of all in Szalatnak-3 borehole. Classification of the Formation:

a) *Upper Siliceous Shale Member.* Dark grey, sometimes black coloured siliceous shale beds alternating with light grey tuffite streaks of about 110 m thickness. On the uppermost part about 20 m thick reddish brown Premesozoic oxidation zone can be found.

b) *Agglomerate Member.* It consist mainly of diabase, andesite, porphyrite, diorite and granodiorite as well as metasandstone fragments embedded in dark grey coloured coarse grained tuffaceous sandstone matrix. It measures about 80 m thickness. The boundaries of the Member are not sharp.

c) *Lower Siliceous Shale Member.* Lithologically it is similar to the Upper Siliceous Shale Member. Certain difference can be observed between two units apart from stratigraphical position of theirs, namely a weak metamorphism in the lower member containing prehnite. The weak metamorphism with prehnite may be regarded as not a Barrow type, but it requires further studies.

Some badly preserved fossils were recorded in the black shale bands in the middle zone of the Member: e.g. *Monograptus* sp., *Hystricosphaeridae*, *Girvanella* and algae by J. ORAVECZ [1964] and M. SIDÓ [1969]. The Lower

Siliceous Shale Member measures about 205 m thickness in Szalatnak-3 borehole.

Precambrian granodiorite-like rock with large K-feldspar phenocrysts underlies the lowermost part of Szalatnak sequence [Gy. WEIN, 1966; GY. BUDA, 1972]. The age of the rock based on radioactive age determination (1100 m.y. by Á. KOVÁCH, [1967] but later it was rejuvenated). According to other authors a narrow contact zone is hidden by strongly fractured rocks between shales and underlying granitoids disturbed by diabasic sills of uncertain age [K. VÁRSZEGI, 1971; B. JANTSKY, 1974]. These authors changed the Precambrian age of the crystalline rocks into Carboniferous in general, and the granodiorite-like rock is considered as being a subvolcanic syenite-porphyr body (laccolite). On the basis of geological point of view, the latter opinion is not fully accepted because of the lack of decisive proofs. Regarding the first opinion of Precambrian age — in spite of more probability of its — it also requires further supports. A new borehole near Szalatnak-3 may gives realistic informations about the lowermost part of the Szalatnak sequence and it could solve the age problems of the Szalatnak Premesozoic formations.

2. *Ófalu Phyllitoide Group*

During the earliest manifestation of Variscan Orogeny an Old-Paleozoic metamorphic belt had wedged along the main axis of the granite range with more than 80 km length and 2–5 km width. Due to a considerable transcurrent faulting the wedged rocks as well as a narrow zone of the country rocks suffered a metamorphism and mylonitisation. The schistosity planes are generally parallel with the bedding planes; — the latter dips are about 330°/54°–80°.

The rocks of the Group are generally covered by Miocene and/or Upper Pannonian deposits except surroundings of Ófalu village (Eastern Mecsek Mts.) where they are outcroping in 5 km in length.

The Ófalu Phyllitoide Group under consideration represented originally a nearly complete single large depositional cycle with basic and ultrabasic volcanics. It can be divided into two formations based on development of theirs:

a) *Goldgrund Formation* can be found along the Goldgrund valley and in Eastern part of the Group. From NW to SE direction it consists of at least four members: a spilite-like metavolcanite, a phyllite an amphibolite and a marble one. M. A. E. GHANEM, L. BARANYAI [1969] were the first interpreters of this volcanism. The members of this Formation have not sharp boundaries between them but there are transitional zones in wich the beds of neighbouring members alternate with each-other. Several beds of some members occur each part of the Formation: e.g. tuff streaks (which in large quantities form the phyllite member) can be found as intercalations in all of members. The phyllite member contains yet great number of metagreywacke beds with characteristic siliceous matrix and appreciable K-feldspar content originated by a later K-metasomatism. The thickness of the Formation reaches 1300 m.

b) *Gründl Formation* forms the NW part of Ófalu Phyllitoide Group and named by Gründl valley in Western section of the investigated area. It measures of more than 200 m thickness, but the covered part of the Formation is possible larger. It consists mainly of basic metavolcanites and associated metasomatites. As a matter of fact, the members of this Formation are

particularly metasomatized metabasalts and mica schists according to M. GHONEIM'S observations.

The Ófalu Phyllitoide Group did not contain any fossils. Based on development and volcanic and metamorphic characters of its it can be dated into (Ordovician)–Silurian period. Based on common lithologic and volcanic character of this Group it seems to be identical with Szalatnak Siliceous Shale Formation and Diabase-Phyllitoide Formation of Balkan Mts. No contradicts of this state the almandine-amphibolite facies of Ófalu Phyllitoide Group as a highest metamorphic degree. The eugeosyncline origin of this Group is assumed by GHANEM–RAVASZ [1969].

3. *Western Mecsek Ultrabasite-Serpentinite Formation*

It consists of two elongated rock bodies of 5 km length and 0.8 km width, enclosed into Precambrian crystalline basement and covered by Permian and Upper Pannonian deposits. Fundamental description of the Formation carried out by T. SZEDERKÉNYI [1962, 1970, 1974].

a) *Helesfa serpentinites and associated rocks* are wedged into the axis of Western Mecsek Permian—Triassic brachianticline (deformed during Upper Cretaceous) in fairly perpendicular position enclosed by strongly sheared porphyroblastic granite (really "augen" mylonite). The latter belonged to the Mecsek Granite Formation. The Western part of this complex is covered by Upper Pannonian sandy deposits of about 100 m thickness, and the Eastern part by undisturbed Lower Permian red sandstones and conglomerates. According to geophysical data and results of two deep drillings, the serpentinite and associated metasomatites settled down as a diapir within a wide gap of the granite, uplifted the covering Upper Pannonian deposits and rolled out the granite country rocks.
Chemical and mineralogical composition of the serpentinite mass shows a rather homogeneous distribution within the body, apart from marginal parts of its which are framed by talc schist selwages. The parent rock of the serpentinite mass was first of all pyroxenite which transformed to serpentinite by shearing, produced lizardite and clinochrysotile composition [J. ERDÉLYI, 1971]. Microstructure of the rock is generally very strongly deformed "mesh" one.

b) *Gyód pyroxenite and associated metasomatites* are settled down in Barrow type metamorphites belonging to the foreground of Western Mecsek Mts. covered by 65 m thick Upper Pannonian conglomerates and sands. Dimension of the rock-mass are almost the same as Helesfa one has, and the position of it is perfectly perpendicular and parallel with the schistosity of Görcsöny metamorphites and it can be regarded as a neck. The main part of the body consists of pyroxenite and weakly serpentinized pyroxenite and serpentinite with laminar texture in all of them, due to lineation of the large clinopyroxenite crystalls oriented to boundaries of the rock body.
According to X-ray investigations, the composition of the pyroxenite are predominantly clinoenstatite and a smaller quantity of olivine, basic plagioclase and magnetite-chromite. The serpentinite consists of lizardite and a small amount of clinochrysotile as well as magnetite [J. ERDÉLYI, 1971]. The microstructure of the serpentinized rocks exhibits an undisturbed "mesh" structure without traces of any shearing or cataclasis. Consequently

it can not be expected diapiric or other type movements within the pyroxenite-serpentinite body.

Three types of altered ultramafic rock-bodies can be distinguished on South Transdanubia: serpentinite diapir at Helesfa, pyroxenite-serpentinite neck near Gyód and serpentinized ultrabasic sill (or lava flow) within Ófalu Phyllitoide Group. This ultrabasic volcanism seems to be of the same age, but in absence of radiometric age determination it can be only presumably placed into Silurian period. The age of the serpentinization can be expected to a very long era from the beginning of the last volcanic events up to present-day at Gyód, but it probably had finished in the Late Paleozoic at Helesfa and Ófalu.

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It seems that our knowledge about Old-Paleozoic of South Transdanubia is first of all restricted to the area of Sout–East Transdanubia, due to industrial (oil, coal and uranium) prospectings. The groundfloor of other parts of the region is covered by rather thick (2000–3000 m) Neogene deposits and therefore is largely unexplored. In the Drava Basin there are scattered data origined from oil prospecting boreholes proving a Barrow type metamorphic basement which sporadically is covered by Upper Carboniferous grey sandstones.

On the Northern side of the so called "Zágráb–Kulcs Lineament" [GY. WEIN, 1967] between Mecsek Mts. and Balaton lake we have unique uncertain occurrence of a slightly metamorphosed grey slate and limestone (Öreglak–1 drilling) which seems to be similar to the adequate members of Ófalu Phyllitoide Group. There are no fossils or else evidences to the real age of these rocks.

EVENTS OF VARISCAN TECTONIC UNIT

1. *Paleohercynian events*

Betveen Silurian and Upper Carboniferous there is no proof of existence of Devonian and Lower Carboniferous rocks in the basement of South Transdanubia, although near Szabadbattyán — in suggesting proximity of the region — can be found a Devonian crystalline limestone occurrence thrusting over the Visean shales and limestones [K. BALOGH, 1974]. It presumably is due to a powerful uplift and fracturing of the region accompanied by hypabissal magmatism produced microgranite and aplite weins as well as K-metasomatism in the older formations. Based on the large hyatus during the period of Early Hercynian movements, it can be assumed that the South Transdanubian region belonged to the "Red Continent". But in the depht very important events had been taking place. The area of Precambrian granites and related metamorphic rocks had suffered the first manifestations of Early Variscan movements (Breton phase) simultaneously with uplift, namely the before-mentioned wedging of Old-Paleozoic overlying rocks to the granite mass and large transcurrent faulting. These events were followed by a second granitization process which produced extremly acidic microgranite and aplite veins (Sudetic phase) intruded mainly to the wedged rocks of Ófalu Phyllitoide Group and Western Mecsek Ultrabasite-Serpentinite Formation.

Southward of the Balaton lake (in 10 km distance) there is another important appearance of the Lower Carboniferous phenomena. A narrow (about 3–4 km width) granite zone — covered by Neogene and Mesozoic rocks, explored a few

deep drillings — runs parallel with the Balaton lake and Mecsek Granite Formation. This granite belt extends to Velence Mts. and the Western border of Hungary. However, opposite to Precambrian granites it form intrusion similar to Velence Mts. granite mass [B. JANTSKY, 1974]. The age of the intrusion is placed to Sudetic phase of Hercynian Orogeny based on radioactive age determinations. The range of this narrow granite belt and the belonged country rocks form the core and axis of a long anticline structure which separates the area of South Transdanubia from Transdanubian Middle Mountains (Bakony etc.).

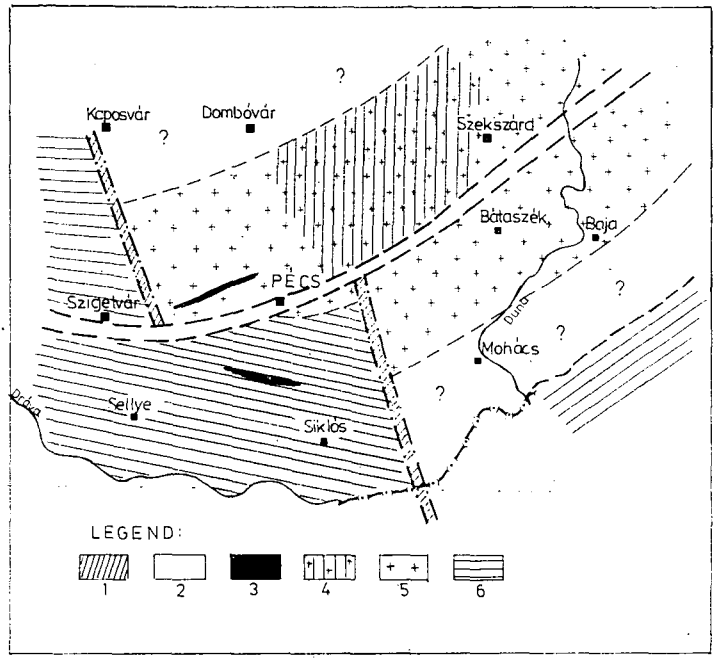

Fig. 1. Geological-tectonic sketch of South-East Transdanubia at the beginning of Late Paleozoic, by T. SZEDERKÉNYI, 1970. Legend: *1.* Precambrian(?) tectonic belt. *2.* Bretonian tectonic belt. *3.* Ultramafic rocks (volcanic vents). *4.* Silurian rocks underlying with granites. *5.* Granites, migmatites. *6.* Crystalline schists with effective strikes.

On the interanticlinal area (between Carboniferous and Precambrian granite range) a connecting link between Paleozoic sequences of Bükk Mts. and Dinaric region by so called "Igal trough" is imagined. There is no aim of this paper to discuss of this conception, but it should also be noted that it is based upon an unique uncertain datum of Karád–1 borehole. By all means the theoretical possibility of Lower Carboniferous sedimentation on the interanticlinal area and the existence of a Paleohercynian trough were rather improbable.

The *Fig. 1* illustrates a tectonic sketch map of South–East Transdanubia at the beginning of the Late Paleozoic after the effect of very first Variscan movements (transcurrent faulting). This aspect is characteristic of all South Transdanubia in this time and shows the connection between Pannonian Mass and a marginal portion of the crystalline basement of Dinaric Geosyncline. The Western part of the map

shows a NW–SE striking belonging to Dinaric type and the Eastern one with SW–NE strikes to the Pannonian type. The northernmost extension of Dinaric structural characteristics can be supposed up to the above-mentioned Lower Carboniferous intrusive granite range.

2. *Neohercynian events*

In contrast to the aforesaid Caledonian and Paleohercynian events, the Neohercynian ones are much more particularly investigated and elaborated, first of all in South–East Transdanubia due to uranium prospecting. There are satisfactory publications available concerning not only Permian of Mecsek Mts. but all over the Hungary. Therefore this chapter of the paper deals with the summary of different interpretations and tries to give some criticism about them in respect to the paleogeography.

It is a well known fact that there is a large scale — at least 4000 m thick — continental series in Mecsek and Villány Mts. put in Upper Carboniferous and Permian period. The study of idealized rock column *(Fig. 2)* reveals some impor-

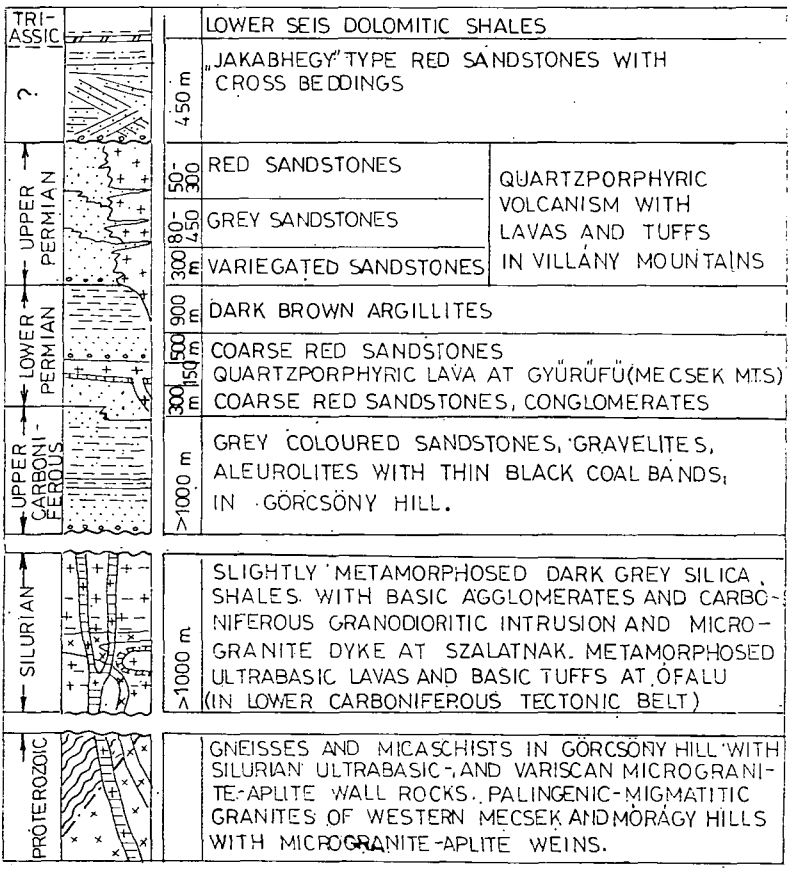

Fig. 2. Idealized rock-column of Paleozoic of South-East Transdanubia, by T. SZEDERKÉNYI, 1974

tant differences between Late Paleozoic of Mecsek and Villány Mts. First of all — according to present-day knowledge — the Carboniferous formation is missing from the sequence of the Mecsek Mts. and is lacking the Upper Permian quartz-porphyric volcanism in the Mecsek Mts. etc. Moreover lateral alterations of thicknesses of the Upper Permian members show a fairly big possibility of error in prediction to a larger distance. In spite of very big density of the data there are controversies about transport directions of the Upper Permian deposits: e.g. based on crossbedding evaluation, those can be originated from NW direction [J. SZABÓ, 1962], but according to the well-founded measurement on lateral alterations of the grain-size, — from the East [M. KASSAI, 1972]. It was not by chance of the first paleogeographic experiment — extended to all of Western Hungary — based on tectonic consideration referring to the Upper Carboniferous and Permian together starting from effectively observed and calculated data [M. KASSAI, 1970, 1972]. At first let us see the palaeogeographic sketch of Upper Permian of South-East Transdanubia by M. KASSAI [1970] *(Fig. 3)*. The estimated area can be divided into two parts by a large fracture zone showing marked NW–SE main directions; this is the so called "Villány–Szalatnak Deep Fracture Zone which is partly coincides

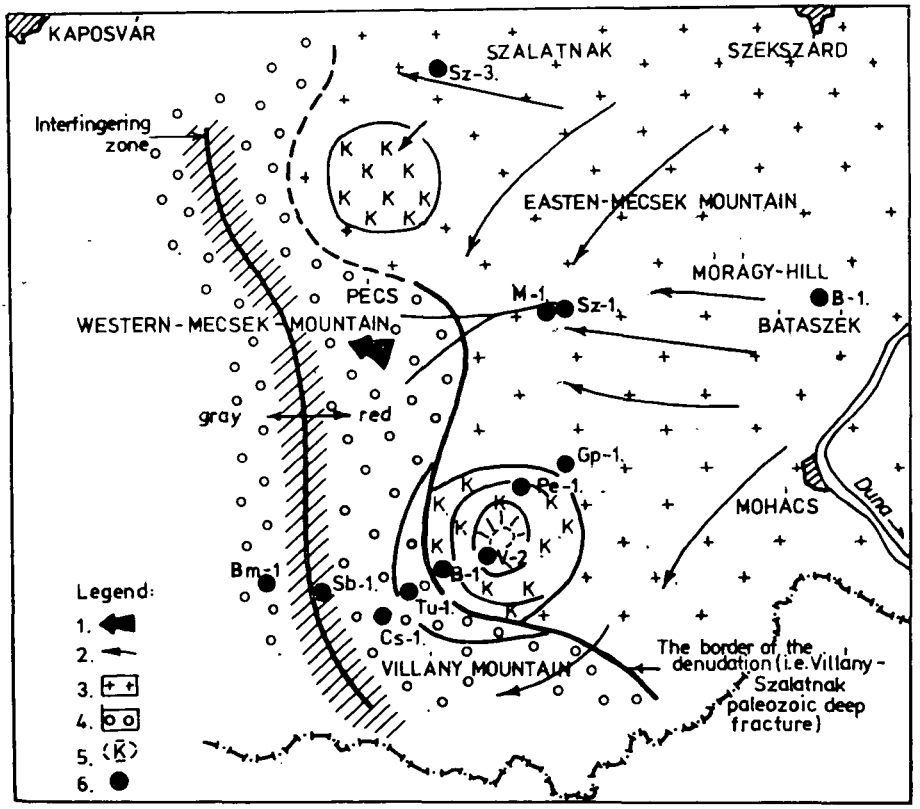

Fig. 3. A palaeogeographic sketch about Upper Permian, South-East Transdanubia by M. KASSAI, [1970]. Legend: *1.* Direction of sediment transport by statistical analysis. *2.* Main direction of deposit-movement on the denudation area. *3.* Denudation area. *4.* Trough. *5.* Quartz-porphyr volcanoes. *6.* Deep drilling.

with the dividing Precambrian (?) tectonic belt between Görcsöny metamorphites and Eastern Mecsek granites (see *Fig. 1*). On the Western side of the deep fracture zone there was a trough filled up with Upper Carboniferous and Permian deposits, and a denudation area on the opposite side with deposit-production for the trough. In the author's interpretation of this sketch, the border between basin and denudation area corresponds to the above-mentioned deep fracture zone, lived during Upper Carboniferous and Permian period, stamped by powerful quartz-porphyric volcanism in the Upper Permian. The lateral distribution of the grain-size and occurrence of an important interfingering zone in the Upper Permian sandstones, show an acceptability of this palaeogeographic picture, nevertheless it would be necessary to confirm it on a few cardinal places. M. KASSAI, [1972] considers this palaeogeographic model valid of Transdanubia in general, and he presents some very thought-provoking proofs to support his conception (see *Fig. 4*). The cardinal place to verify the validity of this idea is the area of supposed "Igal trough".

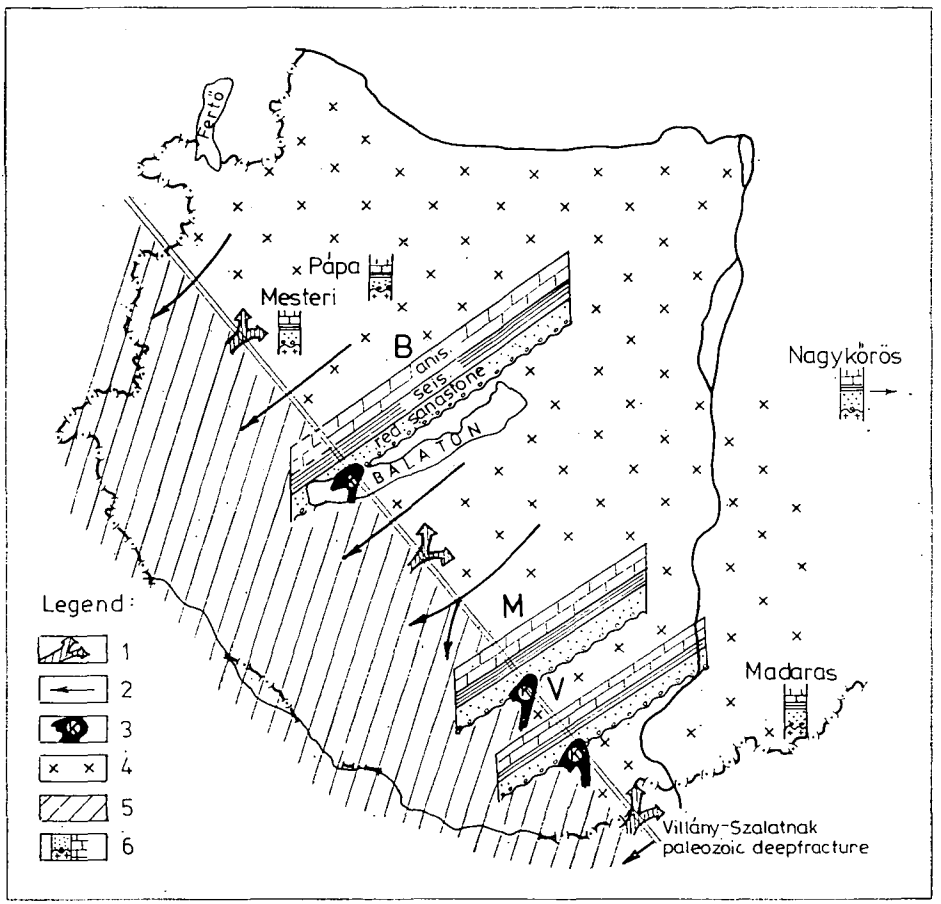

Fig. 4. A palaeogeographic sketch about at the end of Upper Permian of Transdanubia with some typical Lower Triassic profiles, — by M. KASSAI [1972]. Legend: *1.* Direction of Triassic transgression. *2.* Direction of sediment-movement at the end of Upper Permian. *3.* Quartz-porphyric volcanism (Permian). *4.* Area of denudation until end of Upper Permian with crystalline basement. *5.* Upper Permian — Permian trough. *6.* Deep borings.

In the introductory part of this paper the unacceptability of Karád–1 deep drilling was mentioned. The explored and examined rock-material does not belong to the basement. From the debris the Upper Carboniferous age was determined by L. MAJZON, [1966] based on foraminiferas. Apart from the unsuited rock samples of Karád–1 deep drilling, there is a serious failure concerning Upper Carboniferous microfauna of Hungary. Because of more and more species turning into precarious, it is necessary to re-evaluate the Hungarian Paleozoic microfauna. It is easy to see that first of all one ought to make order among obscure originated or misinterpreted data. After all one ought to drill a few boreholes on the cardinal points of the "Igal trough", because this is the key of both hypotheses.

A general sinking had taken place at the end of Permian period all over Hungary producing a typical crossbedded red sandstone formation (except Bükk Mts.) with ground conglomerate overlying the Paleozoic rocks, or crystalline basement. In diastrophic view this is the introductory formation of the Mesozoic sequence of Hungary. In spite of this general statement there are serious correlation problems between red sandstone of Balaton Highland and Mecsek Mts. which are waiting for a satisfying answer.

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PRELIMINARLY PETROLOGICAL AND GEOCHEMICAL STUDIES OF THE AREA ÓFALU, MECSEK MOUNTAINS, HUNGARY

M. F. GHONEIM and T. SZEDERKÉNYI

SUMMARY

On the northern frame of Mórágy crystalline mass near Ófalu village a detailed petrological investigation was carried out in the Lower Paleozoic sequence. It is named Ófalu Phyllitoid Group dividing into three formations and seven members of alternating eugeosynclinal volcanic and sedimentary association affected a fairly low grade Abukuma type regional metamorphism produced greenschist as well as greenschist-amphibolite transition facies. The volcanism is represented by andesitic-basalt rocks with associated tuffaceous derivatives and albite-porphyry as well as serpentinized ultramafic sill-like body intruded the geosynclinal complex. According to comparative petrochemical examinations the andesitic-basalt rocks derived from oceanic type tholeiitic volcanism.

INTRODUCTION

Since the first works made by J. BÖCKH [1876] and S. ROTH [1876] several attempts had performed to interpret the crystalline basement rocks of South–East Transdanubia. Until the beginning of the last decade they were first of all descriptive ones without any petrologic considerations. Because of intense covering of the region by younger complexes the investigations have carried out mainly in the Mórágy Mts and its northern margin where the crystalline rocks are outcroping.

Recently a considerable amount of data are available on Cenozoic and recent volcanic suites giving a better understanding of the association between the chemistry of these rocks and their setting in the framework of the plate tectonic theory [DICKINSON, 1968; HART *et al.* 1970; JAKES and WHITE, 1972]. Plate-tectonic interpretations of Mesozoic ophiolite complex have been published from many parts of the world [BAILEY *et al.* BEZZI and PICCARDO, 1971; MOORES and VINE, 1971; BAILEY and BLAKE, 1974, and others]. Paleozoic ophiolites have similar interpretation [BIRD and DEWEY, 1970; FITTON and HUGHES, 1970; HALLBERG and WILLIAMS, 1972; GALE and ROBERTS, 1974, KEAN and STRONG, 1975; LOESCHKE, 1975, 1976a, 1976b, and others].

Known of all these it may be to do a new and up to date petrologic interpretation of Ófalu schists using the axiom that the present provides a key to the past, comparison of new data of recent volcanic suites and older volcanic ones may provide a clue to the origin of these rocks. The present preliminary work of Ófalu schists is intended to be a contribution to identify the petrological and geochemical characteristics of those rocks, particularly the volcanic suite, bearing in mind recent interpretation placed on other greenstone belts in the plate-tectonic framework then attempt to discuss the volcanic rock series and tectonic setting as a contribution to the question, whether the eugeosynclinal magmatic rocks of Lower Paleozoic of South–East Transdanubia can help to discover plate-tectonic regimes in the rock record of Paleozoic domain.

GEOLOGICAL SETTING AND LITHOSTRATIGRAPHIC CLASSIFICATION OF THE ROCKS

In the southern part of Ófalu village (Eastern Mecsek Mts) there are alternating sequence of eugeosynclinal volcanic and sedimentary associations which have later been regionally metamorphosed ranging from quartz-albite-epidote-biotite subfacies of greenschist facies according to TURNER and VERHOOGEN [1960] up to greenschist-amphibolite transition facies in TURNERS's view [1968]. The rock sequence represents an Abukuma type metamorphism in MIYASHIRO's view [1961].

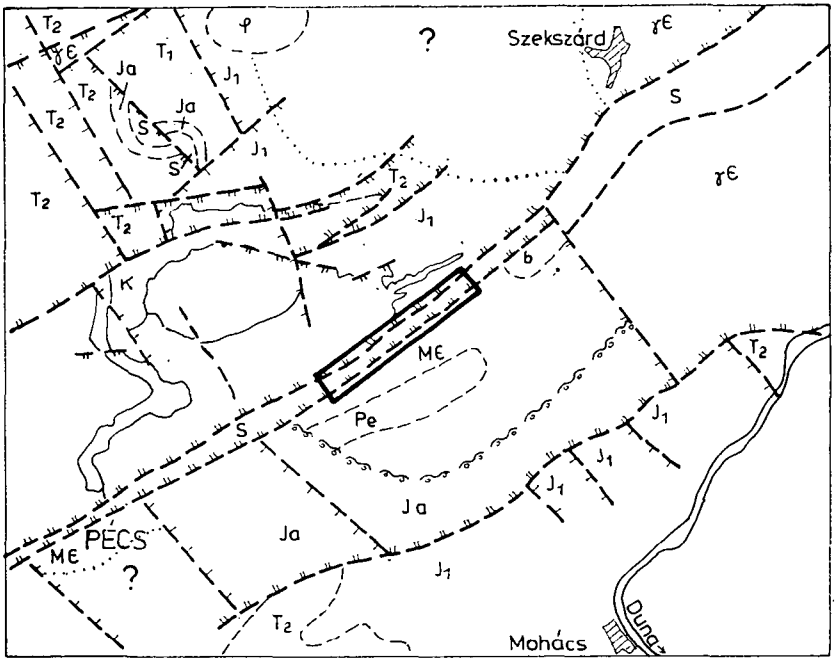

Fig. 1. Site of the Ófalu schists in the groundfloor of South-East Transdanubia.

The following lithostratigraphic classification can be made in the Ófalu schist sequence respected the rules of the International Subcommission on Stratigraphic Classification [1972] and Hungarian Stratigraphic Committee [1975]:

Ófalu Phyllitoide Group	Goldgrung Formation	Albite Porphyry Member Phyllitic Tuffs Member Marble Member Amphibolite Member
	Gröndl Formation	Mica Schist Member (highly metasomatized) Andesitic Basalt Member (metasomatized) Andesitic Basalt Member

This classification is based on petrographic examinations widened by stratigraphic observations.

Andesitic basalts are porphyritic, aphanitic, greyish green coloured rocks. The phenochrysts are plagioclase embedded in a fine grained matrix made of biotite and/or chlorite, few quartz, epidote, sphene, iron oxides and calcite. Plagioclase represents of 30–50% part of the rock; — it has a composition near to peristerite (ranging up to An_{25}).

Chlorite is commonly associated with biotite (together represent about 30% of rock-composition). The analysis of pure chlorite by X-ray diffraction [I. VICZIÁN and M. GHONEIM, 1977] gave a composition close to

$$(Mg_{3,1}\, Fe_{1,5}\, Al_{1,4})\, (Si_{2,6}\, Al_{1,4})\, O_{10}(OH)_8.$$

Biotite is greenish yellow in colour and pleochroic from z=pale brownish yellow or brownish green, x=y greenish yellow. This rock contains some epidote crystalls. The following composition was estimated by electronmicroprobe:

SiO_2	41,4%
Al_2O_3	22,7
FeO	11,7 (total iron)
MgO	0,02
CaO	23,4
	99,2%

Potash feldspars (perthite and orthoclase) are also encountered and they are mainly represents a later origin of potash metasomatism.

Metasomatized andesitic basalts are derived from andesitic basalt by a later metasomatic process which induced potash feldspars and quartz as additional secondary minerals. The essential minerals are highly altered by effect of chloritization, sericitization and kaolinitization. The present rocks are characterized by abundant presence of iron oxides.

Mica schist is used here to denote a regionally metamorphosed and highly metasomatized paste consisting of a mixture of sandy size ($\frac{1}{16}$ mm) quartzo-feldspathic grains and muscovite. Quartz content is abundant, about 20% of the bulk composition. Plagioclase crystalls are predominants suffered a strong cataclasis and alteration. The porphyroclastic "augen" crystal shape rimmed with muscovite, shreds of chlorite and iron oxide are characteristic due to shearing.

Two types of micas are recognized (the white mica and less common mixed layer of chlorite and/or biotite). The white mica was separated and particularly examined by X-ray diffraction. The X-ray studies reveal that the mineral is muscovite in narrower sense having 2 M polytypism with a $\frac{Na}{Na+K}$ of about 4%. The white mica are bended and twisted along the felsic constituents and generally form alignments marking the schistosity planes of the rocks.

Amphibolite microscopically consists of hornblende, plagioclase, chlorite and quartz together with calcite, epidote and iron oxide as accessories. The hornblende is yellowish green in colour and pleochroic from Z=dark green, B=olive green and X=yellowish green. It represents about 30% of the rock constituents. The chemical analysis of hornblende (Table 2) gave a composition close to

$$(Ca_{1.34},\ Na_{0.45},\ K_{0.112})\, (Al_{1.84}Ti_{0.6},\ Fe^{3+}_{1.06},\ Mg_{1.4}/Sr_{1.84},\ Al_{6.09}).\ O_{24}(OH)_{1.49}.$$

Plagioclase represent about 25% of the total volumen of the rock and it has a composition of An_{25-30}. Chlorite (20%) occurs as fibrous aggregates associated with hornblende.

Phyllitic tuffs are fine grained greyish green in colour and have of phyllitic appearance in handspecimens. They consist of oriented fine folias of biotite and chlorite alternating with albite and/or calcareous bands. The volcanic natures of these rocks are represented by abundant presences of albite as the main feldspar mineral, scarcity of quartz crystals and absence of muscovite. Iron oxides are the main accessories.

The marble outcrops measure from 150 to 200 m in thickness. It is light yellow to greyish white in colour and highly massive and brittle rock. Under the microscope the rock consists of mosaic aggregates made of calcite and quartz interrupted by muscovite and limonite. Garnet and diopside are encountered in some specimens.

Albite porphyry commonly intrudes Goldgrund Formation rather than Gröndl Formation, and it is not easily distinguished in the field. The rock is hemicrystalline and exhibits porphyritic texture. It consists of granoblastic albite phenochrysts enclosed in matrix made of chlorite, sericite, quartz and sometimes calcite. Opaque minerals are widely distributed within the groundmass. As it is indicated by X-ray, the quartz represents not more than 5% of the bulk composition of the rock. Chlorite forms the main matrix component.

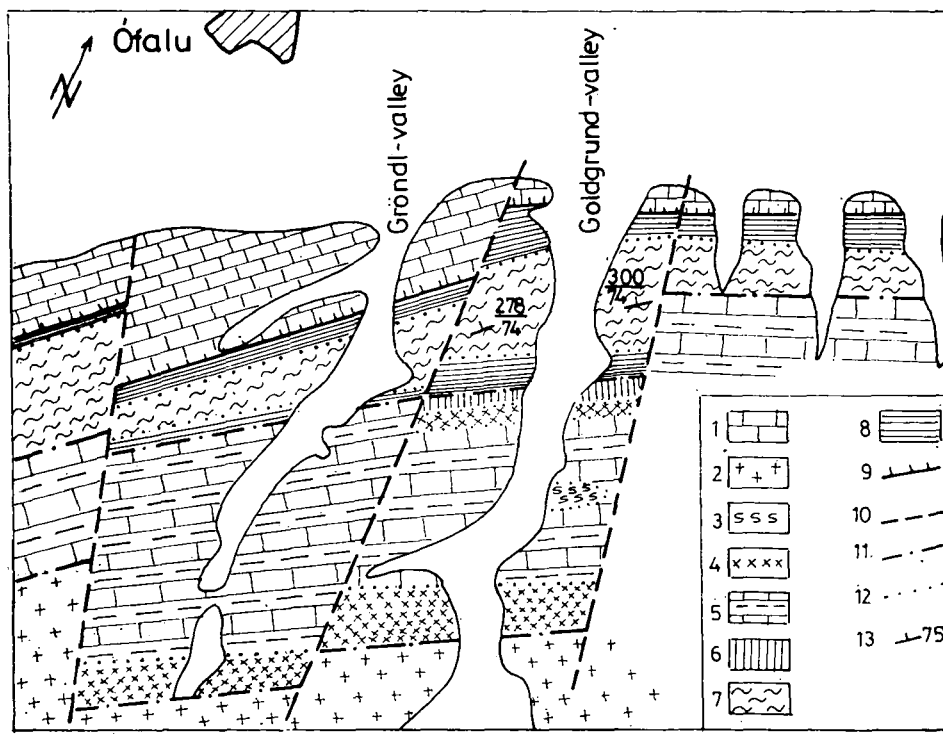

Fig. 2. Geological sketch map of the area Ófalu schists, by M. GHONEIM, 1977. Scale: 1:25 000. Legend: *1.* Jurassic limestone, *2.* anatectic granite, *3.* serpentinite and associated rocks, *4.* albite porphyry, *5.* marble and phyllitic tuffs, *6.* amphibolite, *7.* mica schist, *8.* andesitic basalt and its metasomatized varieties, *9.* Intra-Pannonian overthrusting zone, *10.* faults, *11.* approximate formation contact, *12.* gradational contact, *13.* strike and dip.

The serpentinite and associated varieties wasintruded into the geosyncline in form of sill-like body [GHANEM, R. BARANYAI, 1969] in strict conformity with the regional strikes of the recipient metasediments. Under the microscope the rocks consists of fine aggregates of serpentine minerals together with carbonates, chromite and magnetite. It has a "mesh" texture. Most of the rock specimens contain chromite grains, the rims of which are intensely altered into iron-rich chromite surrounding the original chromite crystals.

PETROCHEMISTRY

As it was mentioned the Ófalu Group under consideration represents Pre-Devonian sequence of metamorphosed eugeosynclinal volcanic and sedimentary rocks. In such a case it is always more difficult to discuss the petrochemistry of the metavolcanic rocks as that of originally magmatic ones, since these rocks take up water during devitrification and metasomatism and change their bulk composition especially those which affected by a later K-metasomatism.

To have highly estimated chemical data as well as acceptable petrochemical characters, the following precaution are taken into consideration:

1. The samples for chemical analysis, were selected to have no secondary potash feldspars as a result of K-metasomatism and minimum alteration. This selection was made by microscope and then checked by X-ray.
2. The most convenient way to identify the present volcanic rock suite is to use elements that show clear chemical differences between the different magma types and are also not affected by secondary alteration processes.

Elements that have undergone little change during such alteration include Ti, Zr, Y, Nb and Sr [CANN, 1970, PEARCE and CANN, 1971] using such "immobile elements" it has been demonstrated that the different magmatypes can be geochemically separated and characterized in altered rocks [FLOYD, 1976].

The major and trace element analysis for 12 selected rock samples of the metavolcanites of the Ófalu Group are represented in Table 1a. The cation mesonorms for these analyses are comprised in Table 1b. In such rocks it is obvious that the calculation reflects the alteration of these rocks which will be discussed later with special care. The number and origin of the samples is the following:

Sa–5	Andesitic basalt (from Goldgrund valley)
203 and 204	andesitic basalt (extreme eastern tributary near Ófalu village)
13	andesitic basalt, (Goldgrund valley, GHANEM and R. BARANYAI 1969)
99	calcareous phyllitic tuff (Gröndl valley)
401753	Albite porphyry (Goldgrund valley)
94	calcareous phyllitic tuff (Gröndl valley)
2	albite porphyry (Grönd valley, GHANEM and R. BARANYAI, 1969)
Sc. 17	Amphibolite (Goldgrund valley)
126	Amphibolite (Studer valley, GHANEM and R. BARANYAI, 1969)
226	Amphibolite (Bátaapáti village)
88/1	Amphibolite (Bátaapáti, JANTSKY B., 1974)
330, 331, 321	Ocean floor type basaltic rocks of Lower Paleozoic Grimeli Formation of the Stavenes Group, Norway. (GRALE, 1975)

TABLE 1A

Major and trace element analyses (wt%) for rocks of Ófalu Group compared with ocean floor-type basaltic rocks of Lower Paleozoic Grimeli Formation of Stavenes Group, Norway [GALE, 1975]

	Sample No.														
	Sa—5	203	204	13	99	401753	94	2	Sc- 17	126	226	88/1	330	331	321
SiO_2	53,12	50,40	52,01	45,50	52,29	64,54	48,01	50,24	49,47	51,64	49,97	49,96	48,00	49,70	49,60
TiO_2	1,26	2,03	1,59	1,59	2,25	1,00	2,45	2,51	1,81	2,44	2,38	1,50	1,92	1,38	2,31
Al_2O_3	18,65	16,59	16,23	18,79	16,06	15,40	13,32	14,87	12,90	13,97	15,19	13,30	13,60	15,20	13,10
Fe_2O_3	2,01	2,83	2,54	3,76	5,36	1,23	1,00	2,40	3,01	3,34	4,13	3,80	13,60+	9,70+	13,90+
FeO	4,46	4,05	3,20	5,35	1,31	3,41	7,66	7,98	10,32	8,10	8,25	6,63			
MnO	0,06	0,10	0,08	0,12	0,15	0,06	0,13	0,21	0,17	0,18	0,22	0,18	0,21	0,17	0,27
MgO	4,71	6,58	4,51	6,81	1,53	2,62	5,46	6,18	7,85	5,80	4,79	6,25	6,900	9,00	6,50
CaO	2,40	5,57	5,18	6,12	6,97	1,79	7,91	6,24	6,55	7,53	9,55	10,02	11,6	10,90	11,20
Na_2O	4,80	4,11	4,80	2,92	7,20	4,08	2,10	3,94	2,38	4,26	2,82	3,96	2,96	3,10	3,00
K_2O	3,09	3,30	3,75	2,50	0,42	2,61	1,38	0,62	0,95	0,52	0,33	0,75	0,14	0,09	0,14
$-H_2O$	0,41	1,14	0,02	0,11	0,04	0,06	0,05	0,11	0,22	0,09	0,27	2,02	1,50++	2,34++	1,76++
$+H_2O$	3,31	2,33	2,94	4,91	1,48	2,47	4,57	3,01	3,32	1,99	2,24	0,14			
CO_2	0,52	0,98	1,96	0,92	4,47	1,01	5,40	0,27	0,59	0,03	0,11	1,62	—	—	—
P_2O_5	0,90	0,80	0,90	0,29	0,47	0,16	0,39	0,48	0,18	0,23	0,45	0,15	0,11	0,07	0,13
Cr_2O_3	0,00	0,01	0,01	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	—	—	—
Total	99,98	99,80	99,67	99,69	99,24	100,4	99,83	100,0	99,73	100,1	100,7	100,3	99,74	101,6	101,9
The analyses of Hungarian samples made by M. EMSZT															
Zr	1000	1800	650	1150	250	120	200	180	200	145	250	—	151	99	185
Sr	2000	3900	2000	5700	165	230	250	128	130	280	260	—	164	135	128
Rb	31	20	28	N. D.	14	N. D.	N. D.	N. D.	N. D.	N. D.	N. D.	—	0	0	2
Zn	75	95	55	67	48	55	250	120	90	65	3200	—	98	60	112
Ni	70	70	N. D.	N. D.	45	38	50	N. D.	65	N. D.	N. D.	—	61	113	56

Trace element analysis (in ppm) by X-ray fluorescence method made by G. RISCHÁK
N. D. = Not detected
\+ = Total Fe content in Fe_2O_3
++ = Total H_2O content
— = Not analyzed

TABLE 1B

Cation mesonorms of the rocks of Ófalu Group

Sample No. / Miner.	Sa—5	203	204	13	99	401753	94	2	Sc. 17	126	226	88/1
Q	12,00	7,70	7,23	6,39	9,96	29,27	20,70	2,93	7,88	8,63	5,08	5,16
Ab	44,00	37,30	43,50	27,35	63,95	37,50	19,85	37,05	22,50	39,20	26,40	36,00
An	—	9,65	1,40	17,95	—	—	—	18,65	—	7,75	5,94	0,80
Or	2,88	0,48	9,33	—	—	12,30	—	—	—	—	2,05	4,50
Bi	25,31	30,65	21,00	25,64	3,93	8,62	13,58	6,08	9,47	5,04	—	—
Ho	—	—	—	—	—	—	—	—	34,00	26,19	46,80	40,36
Hyp	—	—	—	7,48	1,26	—	17,70	24,18	12,48	0,10	—	—
Ap	1,90	1,62	1,93	0,61	0,88	0,33	0,93	0,93	0,48	0,48	1,16	0,29
Sph	1,77	4,20	3,36	3,48	2,01	0,99	2,10	5,43	4,02	5,22	5,22	3,21
Cal	1,37	2,46	5,06	1,22	11,24	2,60	13,90	0,70	1,32	0,08	0,34	4,16
Mt	2,13	2,94	3,20	4,05	3,00	1,31	1,05	2,61	3,33	7,14	4,51	4,05
Rut	0,41	—	—	—	0,87	0,37	—	—	—	—	—	—
Cor	8,23	2,99	4,32	5,68	4,10	6,64	9,59	1,37	4,37	—	3,17	—
Hem	—	—	—	—	1,70	—	—	—	—	—	—	—
Total:	100	99,99	100,3	99,85	99,90	99,93	99,40	99,93	99,85	99,83	100,7	99,03

Major element analysis of hornblende separated from amphibolite rocks, Goldgrund Valley, Ófalu

TABLE 2

SiO_2	45,97%
TiO_2	4,24
Al_2O_3	11,71
Fe_2O_3	5,98
FeO	9,46
MnO	0,20
MgO	7,58
CaO	8,34
Na_2O	1,62
K_2O	0,69
$-H_2O$	0,12
$+H_2O$	3,16
CO_2	0,44
P_2O_5	0,09
Total:	99,60%

Analyzed by M. EMSZT.

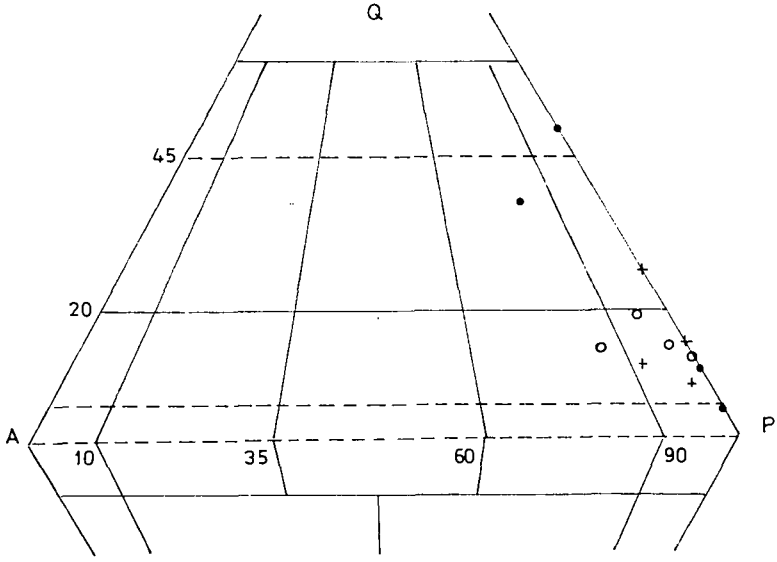

Fig. 3. Distribution of the Ófalu metavolcanics in the QAPF diagram [STRECKEISEN, 1967]
O andesitic basalt
+ amphibolite
• albite porphyry

The cation mesonorms are represented on QAPF diagram [STRECKEISEN, 1967]. This figure nicely reveals that the metavolcanics of the Ófalu Group were originally andesitic basalt and plot mainly in the basalt field of QAPF diagram (some analysis were plotted outside these fields probably due to secondary alteration).

Then the next step in this work is to identify the type of basalt included in the Ófalu Group by help of MIYASHIRO system.

MIYASHIRO [1975a, and b] had classified the volcanic rock series of the Earth into two main groups: alkalic and nonalkalic ones. The latter may by subdivided into tholeiitic (Th) and calc-alcalic (CA) series. For the distinction between alkalic and nonalkalic series, the HARKER's-type variation diagram with SiO_2 content on the abscissa [HARKER, 1909] was adapted particularly by MACDONALD and KATSURA [1964] and then MIYASHIRO [1975a].

Illustrating of Na_2O+K_2O versus SiO_2 with KUNO's boundary *(Fig. 4)* reveals that Ófalu metavolcanics contain representatives of both alkalic and tholeiitic basalt magma series. The metavolcanic rocks of the Gröndl Formation are represented in the alkali field, while the metavolcanic members of the Goldgrund Formation are mainly in the tholeiitic basalt field of division.

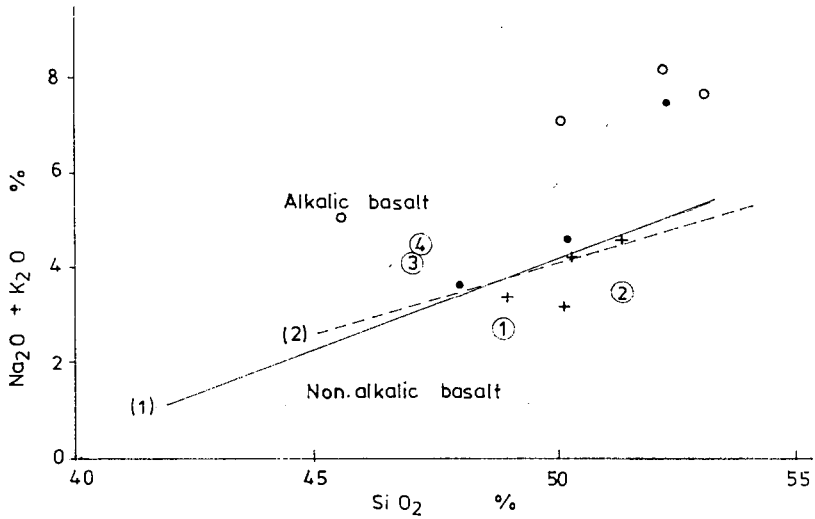

Fig. 4. Alkali-Silica variation diagram. The curve (1) means the boundary between the alkalic and tholeiitic basalt in Hawaii [MACDONALD and KATSURA, 1964] and curve (2) gives the boundary between alkalic and nonalkalic volcanic rocks in Japan [KUNO, 1966]. Symbols:

O andesitic basalt from Ófalu
\+ amphibolite from Ófalu
· albite porphyry from Ófalu
1 oceanic tholeiitic basalt [MANSON, 1967]
2 continental tholeiitic basalt [MANSON, 1967]
3 oceanic alkalic basalt [MANSON, 1967]
4 continental alkalic basalt [MANSON, 1967]

In fact this plot is not entirely reliable in this context. The unreliable nature, particularly of the alkalic series is due to variable migration of akalics during metamorphism as well as the role of secondary alteration and interaction with foreign material, the latter will be discussed in another proceeding work.

The chemical data of the metavolcanics of the Goldgrund valley are presented in standard AFM diagram *Fig. 5*. The data plot firmly within the tholeiitic field of composition defined by IRVINE and BARAGER [1971].

For graphical distinction between tholeiitic and calc-alkaline series of basalt, MIYASHIRO [1973 and 1975b] had proposed the use of variation diagram based on variation of SiO_2, TiO_2, FeO wt. percent (where FeO means total iron as FeO) with

advancing fractional crystallization (the latter is indicated by FeO/MgO ratio). From *Fig. 6a, b, c*, it can be seen that the metavolcanic rocks of the Goldgrund Formation tend to plot well within the fields of the tholeiitic rock series.

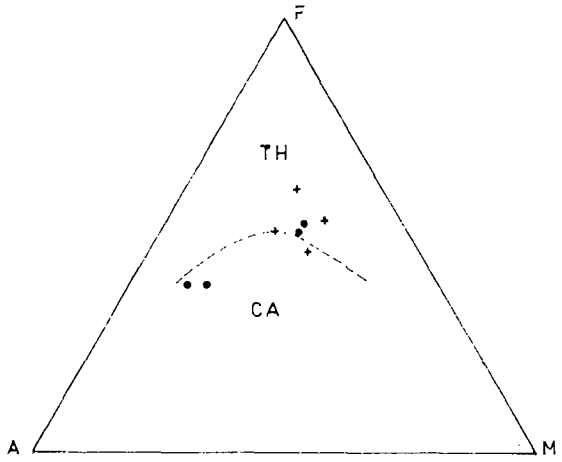

Fig. 5. A=(Na_2O+K_2O); F=(FeO, total Fe); M=(MgO) variation diagram. The field boundary separating tholeiitic (above) from calc-alkalic composition. After IRVINE and BARAGER [1971]. Symbols:
+ amphibolite
· albite porphyry

TECTONIC IMPLICATION OF THE ÓFALU THOLEIITIC BASALT

There is close relationship between volcanic rock series and tectonic setting of theirs. MIYASHIRO [1975a, b] reported about the abundance of the volcanic series on the Earth. Regarding the tholeiitic series they are predominant in midoceanic ridges and other plateau basalts, and they are the most abundant volcanic rocks on the Earth. Although tholeiitic basalts are mainly originated at the oceanic ridges, the continental basalts of tholeiitic nature cover large areas.

PEARCE and CANN [1973] compiled a large number of analytical data of oceanic and continental tholeiites. By means of a TiO_2—P_2O_5—K_2O diagram were able to discriminate the two types. The tholeiitic rocks of Ófalu Group are plot in this discrimination diagram well inside the field of oceanic tholeiites *(Fig. 7)*.

A method using the trace elements Ti, Zr, Y, Sr and Nb as discriminants in distinguishing the tectonic environment of generation of basic volcanics has recently been outlined [PEARCE and CANN, 1971, 1973, GALE and ROBERTS, 1974, and FLOYD, 1976]. The elements Ti, Zr, Y and Nb do not appear to be mobile under conditions of greenschist facies metamorphism, whereas Sr can slightly affected by secondary processes. The different tectonic environments of magma generation can be determined by Ti—Zr and Ti—Zr—Sr plots.

The application of this method to the tholeiite basalt of the Ófalu (Group *Fig. 8* and *9* confirm the presence of ocean-floor basalt, since they plot mainly within the field C in *Fig. 8* and near to the field A of *Fig. 9*. The latter behaviour had been recorded for the ocean-floor basalt from the Lower Paleozoic Grimeli Formation of the Stavenes Group, Norway, by GALE [1975] (Table 1 and *Fig. 9*).

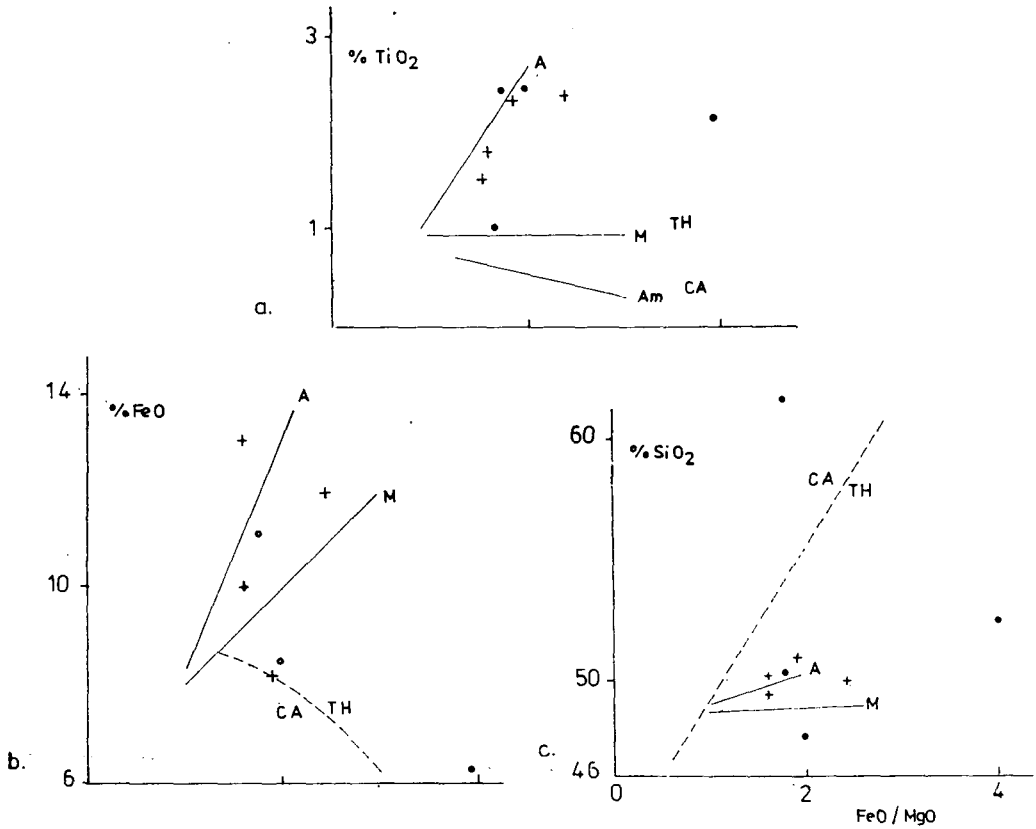

Fig. 6. TiO_2—FeO/MgO and SiO_2—FeO/MgO variation diagrams. The field boundaries separating the tholeiitic (TH) and calc-alkaline (CA), and the trend line for abyssal tholeiites (A), Macauley Island arc tholeiite series (M) and the Amagi calc-alkaline series (Am), after MIYASHIRO [1975b]. Symbols:

+ amphibolite
· albite porphyry

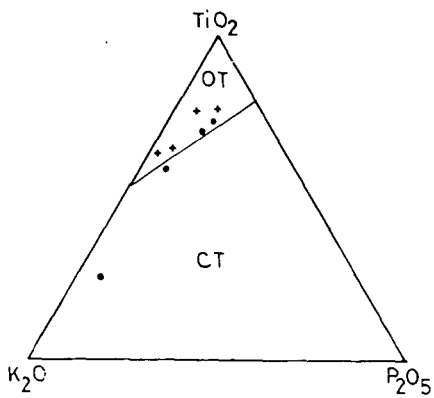

Fig. 7. TiO_2—K_2O—P_2O_5 discrimination diagram with the field boundary of the ocean tholeiite (OT) and continental tholeiite (CT) [PEARCE and CANN, 1973]. Symbols:

+ amphibolite from Ófalu
· albite porphyry from Ófalu

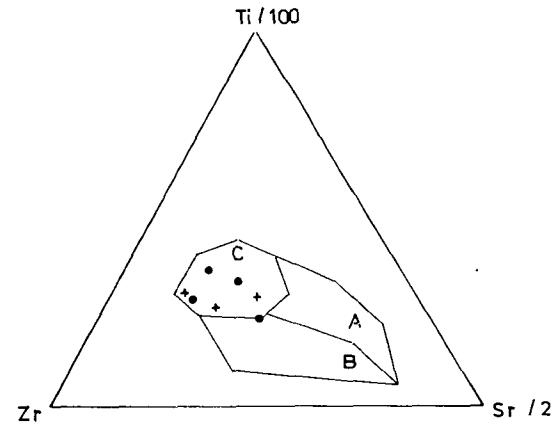

Fig. 8. Ti—Zr—Sr discriminant diagram for distinguishing ocean floor type basalts (field C), low K content tholeiites (field A) and calc-alkaline basalts (field B). After PEARCE and CANN, 1973. Symbols:
+ amphibolite from Ófalu
· albite porphyry from Ófalu

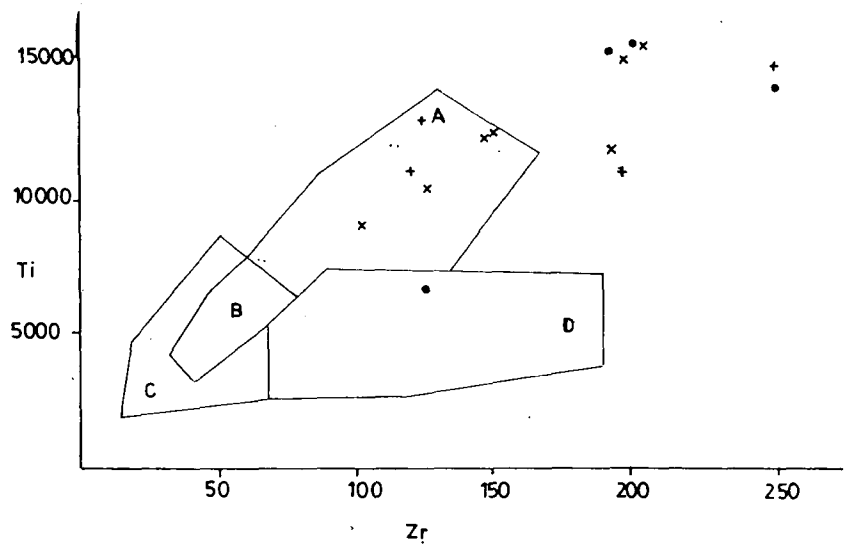

Fig. 9. Ti—Zr plot of Field A=ocean-floor basalts, Field B=ocean-floor basalt and low K content tholeiites of island arcs, Field D=calc-alkaline basalts and andesites. Symbols:
× ocean-floor basalt from Lower Paleozoic Grimeli Formation of Stavenes Group, Norway
+ amphibolite from Ófalu
· albite porphyry from Ófalu

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X-RAY STUDIES ON CRYSTALLINE ROCKS OF THE ÓFALU GROUP, MECSEK MOUNTAINS, HUNGARY

M. A. F. GHONEIM and I. VICZIÁN

SUMMARY

Plagioclase, chlorite, biotite and muscovite component minerals have been separated from the metamorphic rock members of the Ófalu Group, Mecsek Mountains, S-Hungary. The mineral fractions were studied by X-ray powder diffraction methods. The *plagioclases* have composition of (low) albite to oligoclase (peristerites). Chlorites and biotites were separated from greenstones derived from basic volcanic rocks. *Chlorite* formula corresponds to the composition of ripidolites according to the classification of HEY (1954). *Biotites* contain 2.7 (Fe+Mn) and 2.9 (Mg+Al) in six octahedral positions. *Micas* separated from mica schists revealed special characteristics: there is a double 060 reflection at 1.503 and 1.522 Å, respectively, probably due to a mixture of *muscovite with a transitional dioctahedral-trioctahedral type mica* similar to that, which was synthetized by CROWLEY and ROY [1964]. According to the authors' knowledge, no natural analogoues of this letter phase which have been reported so far.

Standard mixtures of the separated minerals were prepared for the purposes of a quantitative X-ray diffraction analysis. Determinative graphs were constructed for hornblende, biotite, chlorite, plagioclase and quartz using metallic Cu as internal standard. Results of the application of the present method for the quantitative analysis of the Ófalu Group greenstones were compared with those of the application of the BRISTOL [1967] method and with chemical mesonorm of the rocks.

INTRODUCTION

The Ófalu Group is a sequence of regionally metamorphosed eugeosynclinal volcanic and sedimentary rocks [SZEDERKÉNYI and GHONEIM, 1977]. The grade of metamorphism ranges from the quartz-albite-epidote-biotite subfacies of greenschist facies [TURNER and VERHOOGEN, 1960] up to the greenschist-amphibolite transition facies [TURNER 1968]. The metavolcanic rock members included in the Ófalu Group are greenstones derived from a basic volcanic suit of andesitic basalt to basaltic composition. The greenstones alternate with metasedimentary mica schists. It was already stated by GHANEM and RAVASZ—BARÂNYAI [1969] that the crystalline schists ot the area "seem to derive from mostly basic products of an initial magmatism connected with the ancient geosyncline".

Aim of the present study is to apply some current techniques of the X-ray diffraction analysis to the mineralogical characterization of the component minerals of these metamorphic rocks as well as to develop and apply a quantitative method to the X-ray determination of the composition of the rock samples. This work represents a part of the detailed petrographical investigation of the Ófalu Group [GHONEIM, 1977].

X-RAY IDENTIFICATION AND CHARACTERIZATION OF MINERALS SEPARATED FROM THE ROCKS

Methods of separation and X-ray diffraction analysis

For a detailed X-ray characterization of the individual mineral phases as well as for the preparation of standard mixtures to the purposes of the quantitative analysis, pure mineral phases were necessary.

The selection of rock samples for separation was preceded by a microscopic and X-ray diffraction analysis in order to check the amount and alteration degree of the component under consideration. By means of a preliminary qualitative X-ray analysis of the individual rock types the following minerals were found (components separated during the present work are italicized):

— in greenstones: *plagioclase, hornblende, chlorite, biotite;* a few quartz, epidote, carbonates and secondary potash feldspar (in some exceptional cases),

— in mica schists: *muscovite, quartz, plagioclase,* potash feldspar; a few chlorite and carbonates.

Mineral fractions were obtained with a Frantz isodynamic magnetic separator in conjunction with a separation in bromoform. The most effective grain size varied from one sample to another in the range of 0.3 to 0.06 mm. Plagioclase and quartz were separated from the light fraction while hornblende, biotite and chlorite from the heavy one. Calcite was removed from the light fraction by treatment with 1:1 HCl. Quartz and plagioclase were picked under a petrographic microscope and then checked for the possible presence of impurities. A slide slope of 20° with magnet set at 0.8 amps in the Frantz isodynamic separator were the best extraction conditions for hornblende, biotite and chlorite. Magnetic separation was repeated several times.

Biotite, chlorite and muscovite are adhesive to some extent. This property was used to a further purification by shaking them carefully on a coarse paper.

Oriented preparates for the X-ray analysis were made by filling sample holder and pressing the powder with a glass plate from above. Non-oriented X-ray preparates were obtained by sieving the fine powder of the material on a coarse paper put under the opening of a sample holder and removing the surplus of the material without the slightest pressure on the sample. The sample holder was then turned over and the coarse paper removed. X-ray analysis was carried out with a Mueller Micro 111–Philips type diffractometer using Ni filtered Cu radiation at 26 kV and 36 mA.

Results

Plagioclase

It was attempted to determine the composition of plagioclases on the basis of the separation of the $131/1\bar{3}1$ and $\bar{2}41/\bar{2}\bar{4}1$ reflections, respectively [BAMBAUER *et al.*, 1967]. The following ranges of variation of these characteristic parameters were found (expressed as $2\Theta^\circ$ for $CuK_{\alpha 1}$ radiation):

TABLE 1

Petrographic name	Number of analyses	$2\Theta_{131} - 2\Theta_{1\bar{3}1}$ (°)	$2\Theta_{\bar{2}41} - 2\Theta_{\bar{2}\bar{4}1}$ (°)
greenstones	18	1,0—1,55	1,4—1,7
mica schists	4	1,1—1,3	1,4

All these characteristic values fall into the domain of peristerites of the low-plagioclase series. No exact determination of the anorthite content from powder data is possible in this interval of composition. It can be concluded, however, that it is the range of albite to oligoclase.

Chlorite

Chlorites of the greenstones are IIb monoclinic polytypic modifications [BROWN, BAILEY, 1962], as it is normally found in metamorphic rocks. The octahedral iron content ("heavy atom content") equals 1.5 ± 0.5 as determined by the PETRUK [1964] method considering six octahedral sites. The "degree of asymmetry" of the iron distribution is $+0.1$. Octahedral aluminum contents were estimated from the exact d-positions of the 004 and 005 reflections using the $d/001/ - \mathrm{Al}$ diagram published by BRINDLEY [1961, *Fig. VI. 7*]. The chemical formula of the chlorites of the samples No. 49 and SA–2 obtained is as follows:

$$(Mg_{3,1}Fe_{1,5}Al_{1,4})\,(Si_{2,6}Al_{1,4})\,O_{10}(OH)_8\,.$$

This formula corresponds to the composition of *ripidolites* in the classification scheme of HEY [1954, see BRINDLEY 1961, Table VI. 3].

Biotite

The octahedral ion content of the biotites of the greenstones was determined by a method of FRANZINI and SCHIAFFINO [1965] recommended by RADOSLOVICH [1975]. No individual ions, only groups of ions of similar atomic scattering properties can be determined by this method. The data obtained for the biotite of the sample No. SA–6 are as follows:

$Fe^{2+} + Fe^{3+} + Mn = 2.7,$

$Mg + Al + 2Ti = 2.9$

(for six octahedral positions, supposing that 0.4 octahedral sites are vacant). The accuracy of these figures is slightly less than ± 0.5.

Muscovite + transitional di-trioctahedral mica (?)

White micas were separated from the mica schists (sample No. 78). X-ray data obtained are as follows:

Polytypism: 2M

$d(002) = 9.975 \pm 0.005$ Å

$$d(060) = \begin{cases} 1.503 \pm 0.0005 \text{ Å} \\ 1.522 \pm 0.0005 \text{ Å} \end{cases}$$

KUBLER's degree of crystallinity $= 2.75 \pm 0.25$ mm $= 0.205 \pm 0.015°$ $2\Theta(CuK_{\alpha 1})$

$\frac{I_{004}}{I_{002}} = 0.43$ (I = peak height).

The parameters listed above are widely used for the X-ray characterization of potassic white micas.

The sharpness of the 002 reflection as expressed by the low value of the KUBLER's degree of crystallinity is in good agreement with the metamorphic origin of these micas.

There are however different opinions concerning the crystal chemical and petrological interpretation of the values $d(002)$ and $d(060)$.

$d(002)$ was determined in oriented preparates as the average of the spacings $4.d(008)$ and $5.d(0010)$ measured on 3 parallel X-ray patterns with a correction using the 4.26 Å reflection of low quartz. The values of $d(060)$ were determined in non-oriented preparates on 5 parallel X-ray patterns with a correction using the 1.541 Å line of low quartz.

From a crystal chemical point of view, it seems to be true that no direct determination of the chemical composition of micas from structural data similar to those listed above is possible. This is mainly due to the fact that the various formulas expressing the relationship between cell dimensions and chemical composition, contain too many chemical variables [e.g. "b_0-axis formula", ZUSSMAN, 1967, RADOSLOVICH, 1975].

Petrological experience has shown, however, that a reliable empirical relationship can be found between some structural and chemical variables considering only the range of variation of the chemical composition frequently occurring under normal natural conditions. In addition, X-ray data can be interpreted directly in terms of various characteristics of metamorphism. It was attempted earlier to relate $d(002)$ values to the Na content an $d(060)$ or b_0 values to the octahedral Fe+Mg content of micas. More recent works consider both variables simultaneously pointing out that high contents of octahedral Fe+Mg (*i.e.* phengitic compositions) influence strongly the $d(002)$ — Na relationship [CIPRIANI *et al.*, 1968; KOTOV *et al.*, 1969; MILKEVICH and KOTOV, 1972; DUNOYER DE SEGONZAC and HICKEL, 1972; SEKI, 1973].

Fortunately, the $d(002)$ value of the Ófalu mica (9.975 Å) lies in the range where the influence of the octahedral substitutions is weak: using the diagrams of the authors mentioned it can be concluded that the $\frac{Na}{Na+K}$ ratio of this mica is not higher than 10% (according to the formulas of CIPRIANI *et al.*, 1968: $\frac{Na}{Na+K}=4\%$ considering only the $d(060)=1.503$ Å reflection).

By means of a statistical study SASSI [1972] divided the dioctahedral mica series into the fields of muscovites s. str. and of phengites at $b_0=9.025$ Å, *i.e.* at $d(060)\approx$ ≈ 1.504 Å. According to this boundary, one of the micas under consideration ($d(060)=1.503$ Å) is a muscovite s. str. It is remarkable, however, that its $d(060)$ value falls just into the interval of statistical minimum between muscovites and phengites.

The relatively strong reflection at 1.522 Å *(Fig. 1)* deserves special attention. This reflection cannot be attributed to any impurity found in the separated mica fraction (quartz, feldspars). It was therefore considered to be a 060 reflection of another type of mica present in the same sample. Indeed, by means of microscopic analysis two different types of "muscovite" have been found in these rocks: the first one occurs as well crystallized muscovite sheets from 0.6×0.25 mm to $1.5\times$ $\times 1.0$ mm large, while the other forms cryptocrystalline shreds and flakes [GHONEIM, 1977].

According to the authors' knowledge, no $d(060)$ values near 1.522 Å have been reported so far for natural micas. This figure is substantially higher than $d(060)$ spacings measured in phengitic micas and celadonites (≈ 1.51 Å) and much lower than those found for trioctahedral micas (≈ 1.53 Å). The mineral characterized by this intermediate $d(060)$ value can be interpreted preliminarily as having a transitional

di-trioctahedral composition. KWAK [1971] synthesized micas transitional between dioctahedral and trioctahedral compositions having b_0 parameters occasionally as low as 9.13 Å (corresponding to about $d(060) = 1.52$ Å . These phases, however,

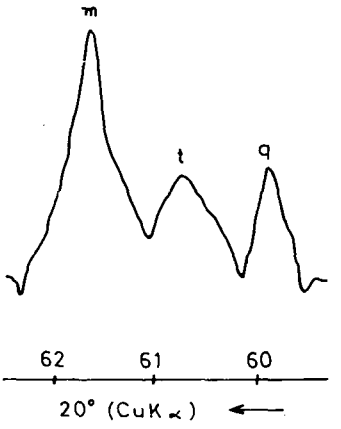

Fig. 1. 060 reflections of the mica fraction of sample No. 78 (mica schist, Ófalu Group). m=muscovite, t=transitional di-trioctahedral mica (?), q=quartz. Conditions of the X-ray diffraction analysis: non-oriented preparate, scanning speed: 1/2°/min, registration speed: 800 mm/h, slits: 1°—0,2—1°, time constant: 8 sec, intensity factor: 1×4

probably have no natural analogues, being pure Fe-Mg-micas that contain no Al. X-ray data of the Ófalu micas agree better with those obtained for some synthetic Mg-Al-mica phases described by CROWLEY and ROY [1964]. According to these data, micas synthesized in the transitional compositional rang ebetween phlogopite and muscovite revealed two distinct 060 spacings almost identical with those found in the Ófalu material (Table 2).

TABLE 2

Material	Reference	$2\Theta(0,60)(°)$ ($CuK_{\alpha 1}$)	
synthetic mica	CROWLEY, ROY [1964, *Fig. 9*]	61,72	60,73
synthetic mica	CROWLEY, ROY [1964, *Fig. 9*]	61,77	60,82
natural mica	Ófalu, present study	61,65	60,79

CROWLEY and ROY considered that the spacings listed above represent two different mica phases each of which being a crystalline solution that "must involve some of the trioctahedral-dioctahedral type substitution" (p. 358). This may be true especially for the phase characterized by $2\Theta(060) \approx 61.7°$.

The unusual "basic" character of these potassic white micas seems to be in accordance with the geological nature of the rock series: it is supposed that the Ófalu mica schists have derived from basic rocks [GHONEIM, SZEDERKÉNYI, 1977].

In addition, however, to this preliminary interpretation, further investigations (e.g. electron microprobe analysis) are needed in order to elucidate the nature of these micas in more detail.

QUANTITATIVE X-RAY DIFFRACTION ANALYSIS

Principles of the method

Unlike sediments, metamorphic rocks are relatively rarely analysed by quantitative X-ray diffraction methods. For the study of the greenstones of the Ófalu Group a method developed by BRISTOL [1967] was adopted with some essential modifications. This is an internal standard method with metallic Cu as reference material. The main deviation from BRISTOL's original method consists in the selection of the components of the artificial "background" mixtures. BRISTOL prepared these "background" mixtures from pure minerals belonging to the same metamorphic facies but taken from different sources while the present authors separated these minerals from the rocks to be analysed.

The importance of the approximate selection of the standard minerals used to the quantitative X-ray analysis has been pointed out by a number of authors dealing with the problem. This is mainly due to the fact that the minerals included into the same mineralogical species or group (e.g.: chlorite, amphibole, plagioclase etc.) may substantially differ from the point of view of their X-ray diffraction intensities [see e.g.: VAN DER MAREL, 1966, RISCHÁK, VICZIÁN, 1974]. This is why it has been frequently proposed to use standard minerals extracted from the rocks themselves that are to be analysed [GIBBS, 1967].

"Background" mixtures were prepared from the components biotite, hornblende, chlorite, quartz and plagioclase. These minerals are present in high amounts in the rocks under consideration. It was attempted to cover the compositional range that may occur in the reality in these rocks. A correction of the "theoretical" composition of these mixtures was necessary in order to take the amount of the impurities of the separated minerals into consideration. Percentages of quartz and plagioclase were determined by a dilution with pure quartz. The actual compositions of the "pure" fractions of biotite, hornblende and chlorite were determined by an approximation using the uncorrected determinative graphs obtained directly from the measurements on our "background" mixtures. These "pure" fractions contained the same minerals that were described in the first part of this work. Hornblende has the formula:

$$(Ca_{1,34}Na_{0,46}K_{0,112})(Al_{1,84}Ti_{0,3}Fe^{3+}_{0,6}Fe^{2+}_{1,06}Mg_{1,4})(Si_{1,84}Al_{6,09})O_{24}(OH)_{1,49}$$

[SZEDERKÉNYI and GHONEIM, 1977].

Methods of preparation and measurement

To 1.00 g of each mixture 0.05 g fine-grained metallic Cu was added. The whole mixture was then homogenized in a rotating mortar in 25 ml of acetone, for about 2 hours.

X-ray preparates were made by filling a Philips rotating sample holder from above and pressing the powder by a glass plate. This preparation is similar to that described by BRISTOL, though producing a somewhat higher degree of preferred orientation. Five parallel X-ray measurements were made for each mixture. Instrument settings were as follows: scanning speed: 1°/min, registration speed: 1600 mm/h, slits: 1°—0.2—1°, angular range: 2 to 50°; $2\Theta(CuK_{\alpha 1}$ radiation).

X-ray intensities were measured as the heights of the reflections above the base line. Base lines were drawn somewhat more flexibly than by BRISTOL (not a completely

straight line across the whole pattern was drawn). This permitted us to obtain zero intensity in each case when the corresponding concentration was also equal to zero. Reflections selected for intensity measurements are listed in Table 3.

TABLE 3

Mineral	d(Å)	$2\Theta°(CuK_{\alpha 1})$	hkl	JCPDS Card No.
Hornblende	8.40	10.52	110	20—481
Plagioclase (low) (Oligoclase)	3.18	28.04	040	9—457
Chlorite (Ripidolite)	7.07	12.52	002	7—76
Biotite	10.1	8.74	001	2—0045
Quartz	4.26	20.82	100	5—0490
Cu (standard)	2.088	43.36	111	4—0836

Results

The measured intensities of the selected reflections of the minerals (Table 3) were divided by the intensity of the 111 reflection of the Cu standard. The intensity ratios $I_{i(hkl)}/I_{Cu(111)}$ were plotted as a function of the weight per cent of the mineral in the standard mixture. Determinative graphs were constructed by " least squares" calculation *(Fig. 2)*. No significant deviations from the linearity could be observed in any case.

The scattering of the points representing the individual measurements around the best fit lines satisfactorily low except for the case of chlorite. This may be due to difficulties of pure separation as well as probably to the different degree of preferred orientation of the preparates containing chlorites.

The agreement with the determinative graphs of BRISTOL is sufficiently good for quartz, biotite and chlorite considering the differences in the choice of standard minerals, methods and apparatus. The graph obtained by us for plagioclase (low) having a composition that corresponds to that of peristerites (albite to oligoclase) lies between the lines proposed by BRISTOL for "albite" and "plagioclase (An_{30})". The largest deviation is that of the graph of hornblende from the line proposed by BRISTOL for actinolite. This may be due to greater differences in mineralogy and grain shape, which are reflected also by the degree of preferred orientation.

APPLICATION OF THE METHOD TO THE STUDY OF THE GREENSTONES OF THE ÓFALU GROUP

The present standard determinative graphs were used for the mineralogical analysis of the Ófalu greenstones. The data obtained by this method were compared with those obtained using the standard graphs of BRISTOL [1967]. Since the determination of the actual modal composition of the Ófalu greenstones by microscopic analysis was not possible because of the fine grain size and high degree of alteration, particularly for chlorite, biotite and hornblende, the X-ray modes were compared with a chemical mesonorm as a reference to the actual percentage of the minerals in the rocks (Table 4).

In chemical mesonorm calculations parts of biotite and hornblende were transformed into chlorite. Because they were difficult to assess the limits of this trans-

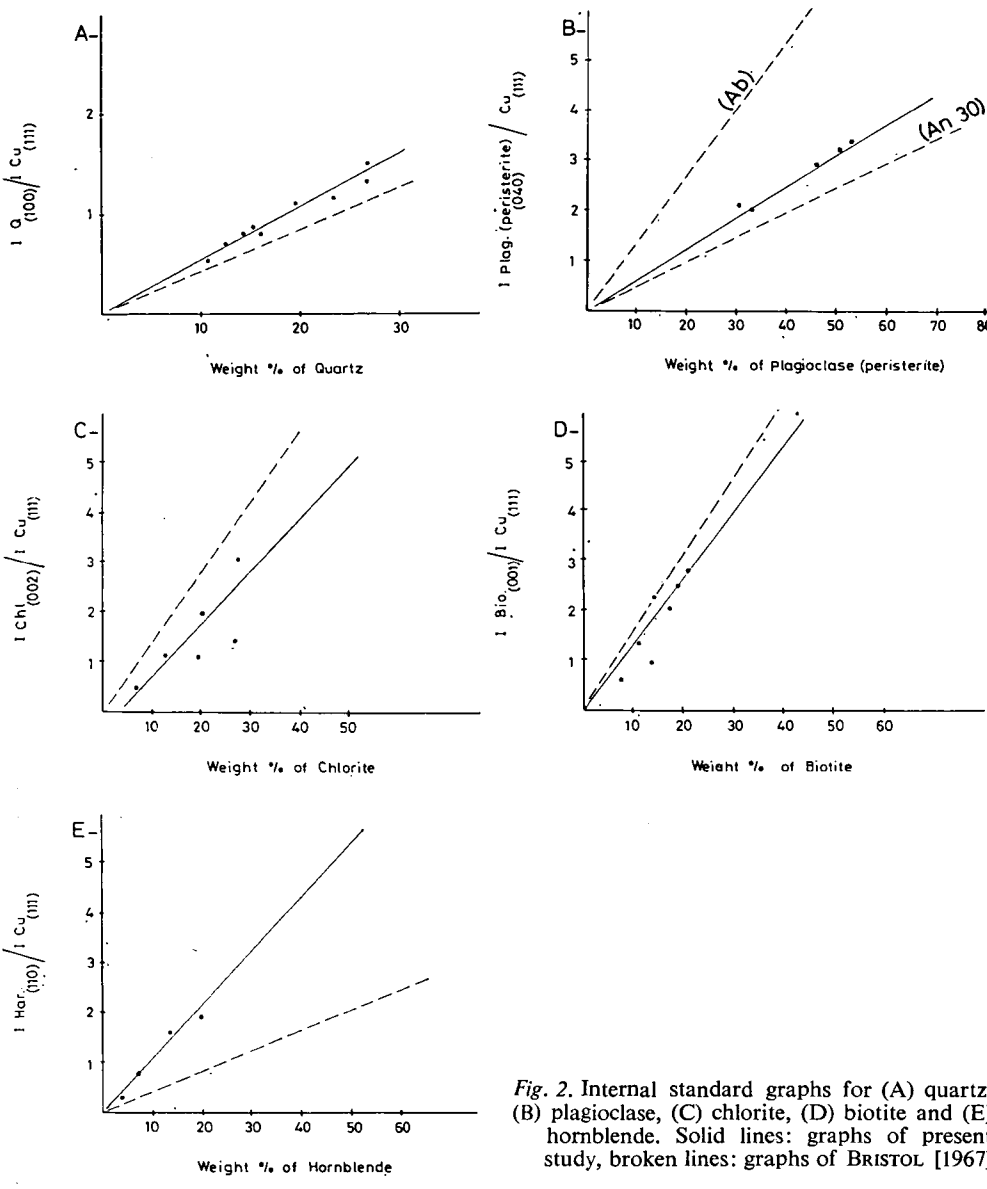

Fig. 2. Internal standard graphs for (A) quartz, (B) plagioclase, (C) chlorite, (D) biotite and (E) hornblende. Solid lines: graphs of present study, broken lines: graphs of BRISTOL [1967]

formation, the samples for chemical analysis were microscopically selected. Three cases can be distinguished:

1. Sample No. Sc. 17 contains a minimum quantity of chlorite and no biotite, but a considerable amount of hornblende.

2. Samples No. 49 and 94 contain a minimum of biotite, plenty of chlorite and no hornblende.

3. Sample No. SA–5 contains equal proportions of biotite and chlorite, but no hornblende.

TABLE 4

Quantitative mineral composition of some Ófalu greenstones
Comparison of data obtained by different methods

Sample No.	Petrographic name	Mineral	X-ray mode		Chemical mesonorm
			BRISTOL's graph	present graph	
Sc. 17	Amphibolite	hornblende	80***	28,5	34,0
		chlorite	10**	17	19,5
		plagioclase	7 (26)*	20	22,4
		quartz	11	8	8,1
49	Andesitic basalt	chlorite	16	20	11,3
		biotite	5	6	5,0
		plagioclase	17 (50)*	39	38,25
		quartz	0	2	3,0
SA—5	Andesitic basalt	chlorite	15	19	13,0
		biotite	11,5	12	8,0
		plagioclase	12 (41,5)*	35	44,0
		quartz	4	5,5	8,1
94	Calcareous phyllitic tuffs	chlorite	15	20	24,9
		biotite	3,5	3	4,0
		plagioclase	6 (25)*	19	19,85
		quartz	17	15	21,5

* In brackets: values calculated from the „plagioclase (An_{30})" standard graph. Values in normal position: calculated from the „albite" standard graph.

** Uncorrected value.

*** Determined using the graph of actinolite of BRISTOL.

In addition to the components listed in the table there are accessories in each sample: calcite, magnetite, sphene, rutile, apatite, epidote, hematite, goethite, orthoclase (secondary).

Table 4 shows that the X-ray modes determined using the standard graphs described here in general agreement with those determined using BRISTOL's standard graphs with some exceptions. As revealed by a comparison of the standard graphs, the largest difference consists in the determination of amphibole (Sample Sc. 17.). The weight per cent of hornblende determined using the present standard graph is nearer chemical mesonorm than so determined as actinolite by BRISTOL's method.

In the quantitative analysis of plagioclases using BRISTOL's method we considered the standard graphs of both "albite" and "plagioclase (An_{30})" since the plagioclase of Ófalu greenstones contains less than 30 per cent anorthite. The values determined with the "albite" graph were far from those calculated by chemical mesonorm. Some difficulties also arose with the use of the standard "plagioclase (An_{30})" graph of BRISTOL (sample No. 49). The data obtained from the present standard plagioclase (albite-oligoclase) graph seem to be near the actual percentages of plagioclase as expressed by the chemical mesonorm values.

It is to be mentioned that details of the calculation of the chemical mesonorm are to be found in the work of GHONEIM [1977].

CONCLUSIONS

In conclusion, it can be stated that the internal standard method as proposed by BRISTOL [1967] proved to be a suitable tool in determining the quantitative composition of metamorphosed volcanic rocks. The published standard determinative graphs, however, either those of BRISTOL, or of the present work, are to be adopted only with great precaution in the case of minerals which may have variable compositions (e.g. amphiboles, plagioclases). The separation of standard minerals from the rocks to be analysed, as described here, seems to be an appropriate way to overcome most of these difficulties.

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DATA FOR A PALEOGEOGRAPHIC RECONSTRUCTION OF TRANSDANUBIA, HUNGARY, AT THE END OF PALEOZOIC TIME

M. KASSAI

SUMMARY

In Mecsek Mts. and Balaton Highland a well studiable characteristic red sandstone group consisting of basic conglomerate with subjacent layers as well as cross-laminated red sandstone, covered by Seisian fauna-bearing formations can be found. In spite of some good criteria of the identifiableness, the conciliatory attempts have produced numerous contradictions during last hundred years. L. LÓCZY sen. and J. BÖCKH had agreed in the stratigraphic identity of both sequence, nevertheless they ranged them into Permian and Triassic, respectively.

E. VADÁSZ had choosen the Permian period, but he did not recognize the character of the main conglomerate of Mecsek Mts. therefore he was unable to do an acceptable comparison between both sequence.

Leaving the lithostratigraphic identity out of consideration lastly K. BALOGH and A. BARABÁS ranged the red sandstones of Balaton Highland and Mecsek Mts. into Permian and Triassic, respectively, similarly to L. LÓCZY Sen. and J. BÖCKH.

The author presents a paleogeographic sketch pointing out possibility of a lithostratigraphic and cronostratigraphic correlation in the mentioned region.

HISTORICAL REVIEW

Studying the history of paleogeographic recognition of Transdanubia regarding the end of Paleozoic, it can be established that there is an abscence of comprehensive appreciation of particular data until the beginning of the last decade.

Among the great predecessors J. BÖCKH [1876] and L. LÓCZY SEN. had agreed in the ability of a comparison between red sandstones of Mecsek Mts. and Balaton Highland covered by Seisian fauna-bearing layers. "The Balaton-side red sandstone and Verrucano can be obviously compared with sandstone of Szentjakab Hill, near Pécs" [L. LÓCZY SEN., 1913]. The stratigraphic relation of them was guaranteed by discordantly settled basis conglomerate as well as even-aged covering strata (fauna-bearing Seisian layers) and that is right at present too *(Fig. 4)*. However both of sandstones were ranged into Triassic by J. BÖCKH, [1876], whereas L. LÓCZY SEN., [1913] had accepted the Permian age for both of them.

E. VADÁSZ, [1912] stated that the Permian sandstone of Mecsek Mts. "...either tectonically or appearance is sharply bordered towards covering formation. Directly above follow banks of the conglomerate discordantly." Consequently he also observed the unconformity of conglomerate. It is very interesting and instructive to follow with attention the change of VADÁSZ's opinion about age of Mecsek red sandstone during a 25 years period. In the Report of Hungarian Geological Institute dated 1917 on the 26-th pages can be red that "...the lower sandstone shows any unconformity towards the conglomerate in most places", but contrasting with this obser-

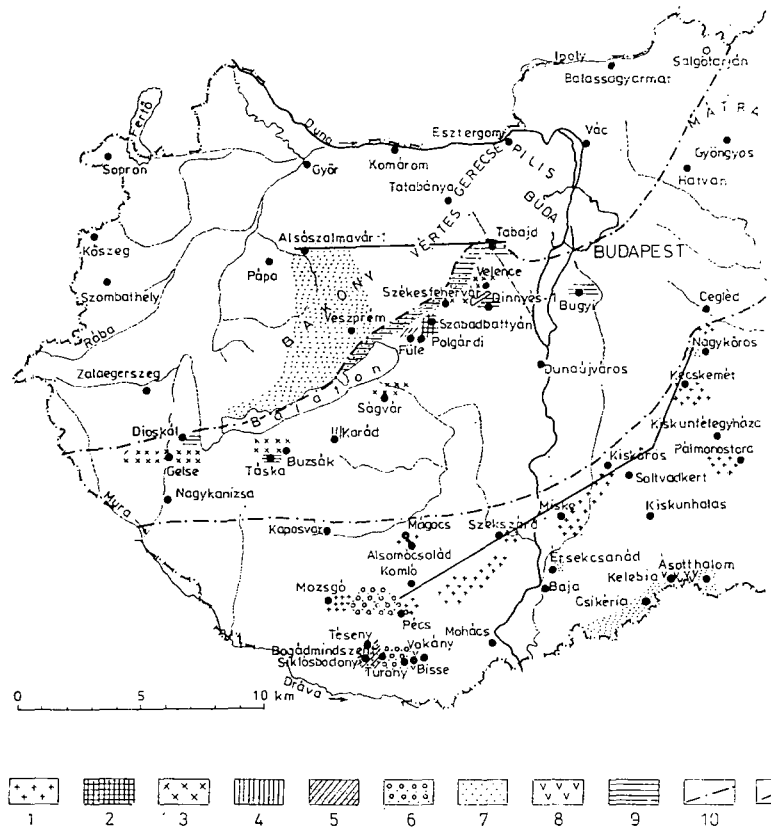

Fig. 1. Review of the occurrences of the Carboniferous and Permian strata and the presumable extension of the sea at the end of the Permian in Hungary by K. BALOGH [1972]. (Part) Legend: *1.* Migmatic granite (Breton phase?). *2.* Early Carboniferous in marine facies. *3.* Granite plutons (Sudetic phase?). *4.* Upper Carboniferous marine facies. *5.* Middle and Upper Carboniferous in continental and marine facies. *6.* Lower and Upper Permian continental facies. *7.* Upper Permian, transgressing over Oldpaleozoic. *8.* Quartz porphyry and quartz porphyry tuff. *9.* Upper Permian in continental-lagoonal, in continental-marine facies, respectively. *10.* Upper Permian shoreline. *11.* Profiles of Fig. 2a, 2b.

vation in monography of Mecsek Mts. produced by E. VADÁSZ, [1935] it can already be seen that "...The sandstone belonging to the lower member... is passing over gradually into coarse conglomerate without any sharp boundary". Referring to the age of the Jakabhegy sandstone (directly above the coarse conglomerate) in one of the same papers from 1917 (p. 390) one can be read that "...it is possible to range the conglomerate and overlying red sandstones either into Triassic or Permian arbitary...". Twenty years later "...the coarse conglomerate and red sandstones represent the Upper Permian epoch..." [1935, pp. 28].

Taking back of the unconformity by E. VADÁSZ had been a serious error involved some trouble in the South Transdanubian Late Paleozoic stratigraphy during a fairly long period. Giving too much freedom for geologists in the judgement of age-questions on the one hand, breaking up certain correlation criteria between Balaton Highland and Mecsek sandstones on the other, which led to a resignation, namely: in spite of

a considerable thickness and development difference it is sufficient to know the existence of Permian strata in both of area and not any more. Due to this chronostratigraphic generalization on the paleogeographic schemes of Hungary made by GY. WEIN [1969] and others, both of area are presented without any difference.

The study of the same topic by K. BALOGH, A. BARABÁS [1972] is already produced any deviation from the above-mentioned view. Although we are obliged to state that the authors used an arbitary ranging instead of systematic analysis of criteria of the correlation. It led to disappearance of the Jakabhegy sandstone from their paleogeographic sketch *(Fig. 1)* because it was ranged into Triassic period. Consequently, it is impossible to draw any acceptable geological profile between Balaton Highland and Mecsek Mts. or Nagykőrös and Mecsek Mts. based on paleogeographic scheme of the authors *(Fig. 2)*.

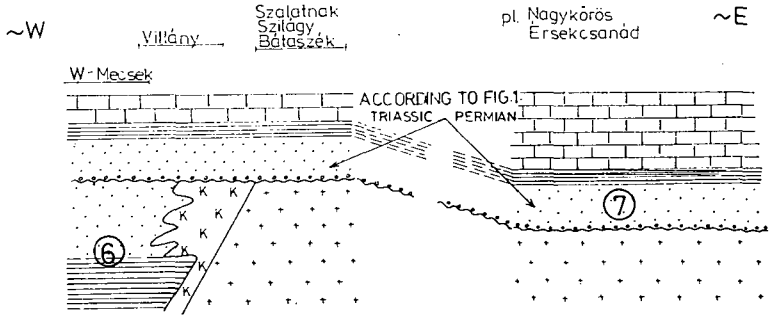

Fig. 2a. Geological profile plotted in *Fig. 1* Legend from the *Fig. 1*

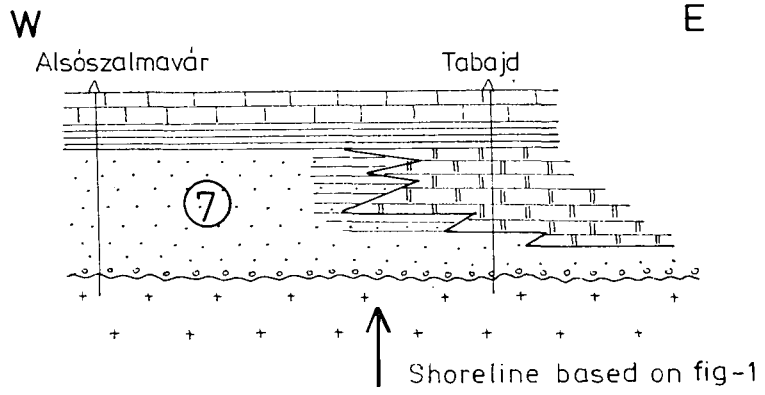

Fig. 2b. Geological profile plotted in *Fig. 1* Legend from the *Fig. 1*

Studied this question historically it should be stated that stratigraphic publications regarding Permian of Balaton Highland since 1913 have been abscence from the Hungarian geological literature.

The paleogeographic reconstruction of Transdanubia in respect to the end of Paleozoic and the beginning of Mesozoic can be carried out well, due to appropriate exploration. The main features of its:

— There was a trough on the western side of so called "Villány–Szalatnak Deep Fracture Zone" filled by a few thousand meters thick Upper Carboniferous-Permian sandstone sequence, while on the eastern side of the fracture zone existed a broad denudation area in the same time [M. KASSAI, 1976].

— With the appearance of the main conglomerate, the eastern area also became trough. Following a sharp uncomformity and showing some tipical stamps of marine transgression, the main conglomerate buried eastward the extinct quartz-porphyric volcanoes and the "old basement". Based on numerous statistical data of the Jakabhegy sandstone it seems belonging to marine main facies developing from the conglomerate and passing over gradually into fauna-bearing Lower Triassic red sandstones and aleurolites [M. KASSAI, 1976].

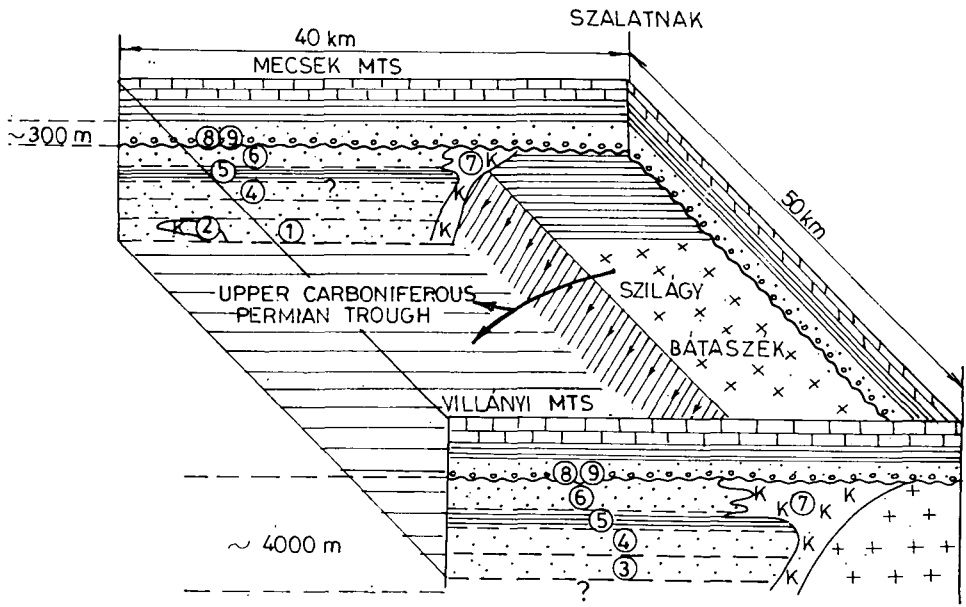

Fig. 3. Paleogeographic setting of the Jakabhegy sandstone of South Transdanubia [M. KASSAI, 1976]. Legend: see *Fig. 4*

This profile is expansible to Nagykőrös boreholes located Duna–Tisza Interfluve without any difficulty. The stratigraphic identity is guaranted by basic conglomerate (main conglomerate) underlying the "old basement" and the superincumbent Seisian fauna-bearing layers as well as identity of the sedimentation.

BALATON HIGHLAND

Used also rock-column descriptions made by K. BALOGH, A. BARABÁS, [1972] it can be established that the red sandstones of Balaton Highland are identical with the Jakabhegy sandstone of South Transdanubia based on well parallelizable underlying and overlying rocks. The basis conglomerate settled down discordantly on the "old basement" rocks similarly to the eastern part of South Transdanubia. This stratigraphic situation as well as regional developmental identity characterize a typical marine transgression which is moreover supported by a general experience, namely: the rivers do not preferre to occupy denudation areas.

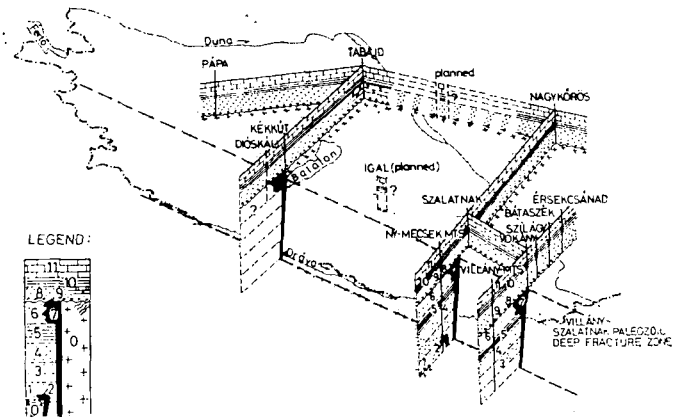

Fig. 4. Geological structure of the Late Paleozoic—Early Triassic in Transdanubia and Danube—Tisza Interfluve. Legend: *1—2.* Gyűrűfű Variegated Sandstone, Congl. and Quartzporphyry Formation. *3.* Téseny Sandstone Formation. *4.* Cserdi Variegated Sandst. and Congl. Formation. *5.* Boda Red Aleurolite Formation. *6—7.* Kővágószőlős Sandst. and Vókány Quartzporphyry Formation. *8—9.* Jakabhegy—Balaton Highland Red Sandstone Formation. *10.* Seisian fauna bearing layers. *11.* Anisian limestone-dolomite

Belonging to the marine main-facies of the red sandstone is proved by heteropic gypsum and marl layers of Tabajd profile. The opinions about continental origin and belonging to the fluviatile main-facies of red sandstones, as well as the statement the "...continental facies in the southern foreland of the Vértes Mountains grades into a gypsum-bearing, lagoonal-marine facies..." K. BALOGH, A. BARABÁS [1972, pp. 201] include some serious contradictions.

AREA BETWEEN BALATON HIGHLAND AND MECSEK MOUNTAINS

There is no an established rock-column available about the area between Balaton Highland and Mecsek Mts. regarding the mentioned period, and this lack of the data can not be substituted by any theoretical consideration.

AGE QUESTIONS

It can be seen from the historical review that chronostratigraphic ranging of the sequences were carried out for themselves, instead of search of the stratigraphic identifiableness criteria. It is a generally known fact that the afore-mentioned sequences

have not acceptable fossils for dating at all, moreover the red sandstones are "totally empty" in fossils.

Its seems to be clear to draw the Permian-Triassic boundary at the fauna-bearing Seisian layers. In relation to this proposition one could remark of two different considerations:

— Drawing the paleogeographic face of this border, it shows a monotonous area consisting of fine grained sandstones, aleurolites and marls. The geologic frame of the Paleozoic trough (which was fairly smaller than Mesozoic one) as well as direction of the Mesozoic transgression, etc. are effaced by this look.

— In many places of the Alpine–Carpathian–Balcan region the red sandstone sequence covering an "old basement", is regarded as the beginning of the large Lower Triassic marine transgression cycle.

Consequently, to the interpretation of paleogeographic setting at the end of Paleozoic, it is necessary a collective presentation of the period ranging from Carboniferous-Permian up to Anisian emphasizing the marks of main lithostratigraphic identifiableness. A standpoint in the age-questions of this topic can be formed only parallelly with the opinions about Alpine-Carpathian-Balcan region.

SHORTNESS IN THE DATA OF PRESENTED PALEOGEOGRAPHIC SKETCH

The presented paleogeographic sketch *(Fig. 4)* contains some lack of the geological data in numerous places. First of all at the marked part of Tabajd–Nagykőrös profile as well as near Igal village can be found cardinal points where from should be gain satisfactory data for an eventual paleogeographic appreciation. In the lack of these data the variegated ideas having a harsh contrast between abundance of them and powerty of the available data, are unacceptable. These cardinal points have also certain economic importance. E.g.: for elaboration of regional hydrocarbon prospecting conditions and conceptions it may be important to know the existence of coal-bearing or organic material-rich Upper Carboniferous and Permian rocks. For the uranium prospecting is outstanding the research of clastic Permian rocks, thus first of all at surroundings of Dióskál, Balaton Highland.

FURTHER CONNECTIONS OF THE PRESENTED PALEOGEOGRAPHIC SKETCH

The presented paleogeographic sketch has appreciable connections within the Carpathian Basin. Same geological rock-columns can be found at Pápa and Little Carpathians as well as Inovec, or Nagykőrös and East- and South Carpathians. It is rightful to look for any connection between Upper Carboniferous-Permian located on the western side of Villány–Szalatnak Paleozoic Deep Fracture Zone and Dinaric Geosyncline as it has already been raised by numerous authors.

Finally it can not be avoided the question of contact with Late Paleozoic development of Bükk Mountains. It is practically insoluble without suggested drillings near Igal and between Tabajd and Nagykőrös.

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METAMORPHIC-GRANITOGENIC ROCKS OF THE BASEMENT COMPLEX OF THE GREAT HUNGARIAN PLAIN, EASTERN-HUNGARY

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ABSTRACT

The metamorphic-granitogenic rocks of the Great Hungarian Plain showing a SW—NE trend have been uncovered by hundreds of hydrocarbon exploring boreholes. The central part is composed of polymetamorphic biotite-plagioclase gneisses, mica schists, amphibolites which gradually pass into migmatites and/or diatexites of granodioritic composition in the marginal parts of the deeply buried ranges involved. The polymetamorphic rocks are intersected by younger granites of medium grain-size. In some boreholes in the southern part of the Great Hungarian Plain mica-quarzites, chlorite schists belonging to the greenschist facies as well as staurolite-mica-schists belonging to the almandine-amphibolite facies have been found, all of them hit by a single metamorphism only. The formation of the metamorphic-granitogenic rocks is a result of three tectono-metamorphic cycles. The first one caused a regional metamorphism of the Proterozoic source rocks into the amphibolite facies and resulted in a synkinematic granodiorite formation. A regressiv metamorphism took place in the second cycle manifesting itself by milonitization and diaphtoresis along narrow zones. The regressive metamorphism resulted in mineral associations of the greenschist facies. These rocks show a limited regional extension until now. The final cycle caused some metasomatic transformations of considerable intensity as well, forming late kinematic granites.

INTRODUCTION

The metamorphic basement complex of the Great Hungarian Plain has been revealed by hydrocarbon exploring boreholes. After having intersected Cenozoic and Mesozoic sequences of different thickness, about 650 boreholes have reached the ancient crystalline rocks. A slightly curving line, connecting Újszilvás–Túrkeve–Hajdúszoboszló–Ártánd exploration areas, can be considered as a boundary north of which the metamorphic basement complex has not been discovered until now.

A comprehensive discussion of these metamorphic formations has been given by three studies [E. SZÁDECZKY-KARDOSS *et al.*, 1967, E. SZÁDECZKY-KARDOSS *et al.*, 1969, K. SZEPESHÁZY, 1973] so far. The petrological, stratigraphic and tectonic characteristics of the metamorphic rocks have been thoroughly discussed in these publications. The connection between the crystallin complexes of the Great Hungarian Plain and those of the Carpathian region has been outlined in the study of K. SZEPESHÁZY [1973]. The development of the metamorphic and granitogenic rocks in the Great Hungarian Plain has been ranged to some tectonomorphic cycles belonging to the Prae-Baykalian, Baykalian and Hercynian orogeny.

The chemical zonality of the garnets of metamorphic origin is considered by P. ÁRKAI *et al.*, [1975] as a proof of the polymetamorphism. By investigating some samples from the Great Hungarian Plain, they have stated that the original zoning of the garnets developed in the first phase of the recrystallization had been covered by a secondary zonal system formed later, during the regressive metamorphism.

METAMORPHIC AND GRANITOGENIC RANGES OF THE GREAT HUNGARIAN PLAIN

The metamorphic-granitogenic ranges forming the crystalline basement of the Great Hungarian Plain show a general SW–NE trend and a symmetrical arrangement. The central — *i.e.* largest — part of the Great Hungarian Plain is composed of polymetamorphic biotite-plagioclase gneisses hit by a regressive metamorphism and of plagioclase bearing mica-schists accompanied by amphibolite masses of local extension *(Fig. 1)*.

The mica-schist-gneisses range passes both towards the NW and towards the SE into granitogenic ranges surrounded by migmatites. The metamorphic-granitogenic basement forming ranges has been intersected by younger, granitogenic formations with SE–NW and/or SW–NE trends. Their common characteristic is that they are narrow, sometime show vein-like nature.

Naturally, the different ranges can not be distinguished without some kind of exploration. The areal distribution of the boreholes reaching the basement is highly uneven; in some areas data of 100 or even more boreholes can be taken into consideration, while in other areas data of some boreholes can be relied only.

Polymetamorphic crystalline rocks

Biotite-plagioclase gneisses

Biotite-plagioclase gneisses have been uncovered boreholes in several areas *(Sükösd, Érsekcsanád, Jánoshalma, Kiskunhalas, Tázlár, Szank, Kelebia, Forráskút, Sándorfalva, Üllés, Dorozsma, Algyő, Ferencszállás, Csanádapáca, Mezőkovácsháza, Végegyháza, Öcsöd, Szarvas, Endrőd, Füzesgyarmat, Komádi, Mezősas, Kőrösszegapáti* and *Ártánd)*.

As a result of a concentrated drilling activity the metamorphic basement complex is much better explored than usual *Jánoshalma–Kiskunhalas–Tázlár–Szank* (Danube–Tisza Interfluve), *Algyő* and in the NW part of the *Mezőhegyes–Mezőkovácsháza* arch (southeastern part of the Great Hungarian Plain).

The biotite-plagioclase gneisses are greenish-gray, medium or fine grained rocks with a schistosity of different intensity. As a result of the strong regressive metamorphism, their original structure had changed i.e. their structure characteristic of gneisses had disappeared and in many cases a secondary schistosity developed.

Their minerological composition is a simple one. The most important primary components are as follow: plagioclase, quartz and biotite. Accessories are garnet and apatite. K-feldspar of primary origin does not occur.

As a result of the regressive metamorphism the sericitization of the An_{28}-oligoclase of primary origin has taken place followed by a decomposition of variable intensity. The intensity of the biotite degradation os somehow different; a number of transitions beginning from the slight chloritization to the formation of chlorite-calcite-magnetite patches can be observed. The mineral assemblage of the regressive metamorphism is composed of chlorite, quartz, sericite, calcite, magnetite. A secondary mineral association consisting of younger K-feldspar, albite, quartz, muscovite frequently emerges and replaces the primary one.

The chemical analyses of the rocks have been summarized in four regional groups according to the possibilities offered by the sampling. The following statistical parameters are given for each rock type and for each area in the Table 1: mean

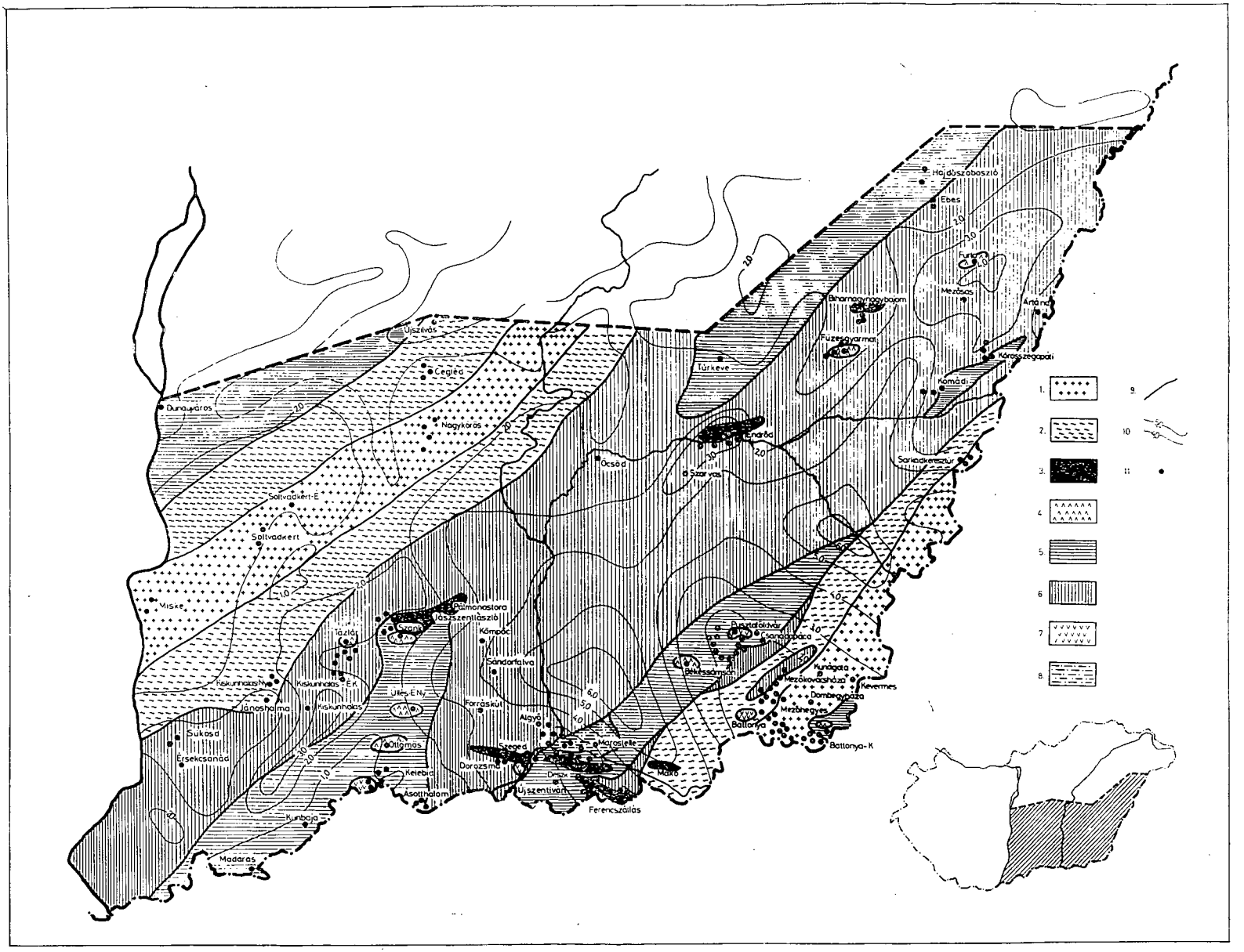

Fig. 1. Metamorphic-igneous rocks in the Great Hungarian Plain
Legend: 1. Granodiorite; 2. migmatite; 3. granite; 4. amphibolite; 5. mica-schist; 6. gneiss; 7. quartz-porphyry; 8. chlorite schist; 9. boundary of the formation; 10. contour on the top of the crystalline basement complex; 11. borehole; 12. boundary of the explored area

value (x), standard deviation (S_x), mean standard deviation ($S_{\bar{x}}$) as well as number of the analyses. In the order to select the different petrological families independently of the sampling areas, STUDENT- (or "t"-) test was done. On the basis of this a correlation can be shown up between the know occurrences of the biotite bearing plagioclase-gneisses. Consequently, the chemical composition of the rocks can be given as a mean characteristic of the whole Great Hungarian Plain (Table 1).

Muscovite-biotite-plagioclase mica-schist

The overwhelming majority of the mica-schists uncovered in the area under consideration are of polymetamorphic nature.

Mica-schists hit by a single metamorphism are know scattered in small patches only. According to our information having acquired until recently, mica-schist-like rocks can be followed in SW–NE trending zones which wedge into or paralell with the central gneiss-range at *Madaras, Kunbaja, Kelebia, Ásotthalom, Algyő, Újszilvás, Ferencszállás, Kiszombor, Pusztaföldvár, Csanádapáca* areas. More scattered occurrences around *Turkeve, Hajdúszoboszló* testify to the existence of a northern mica-schist zone.

The mica-schist-like schists of varying appearance are mostly stressed, folded rocks with greenish shade. In thin sections the following charactereistics can be observed: lamination fabric, unidirectional arrangement of the fairly elongated mafic components, a separation of the mafic and salic minerals in form of systematic alignement of these crystals. The polymetamorphic character of the rocks is referred to by a multi-generation mineral association. Their primary mineralogical composition is rather uniform. The main components are muscovite, biotite, quartz, plagioclase, garnet. Accessories: cordierite, epidote, tourmaline, magnetite, titanite. There are two plagioclase modifications. The original mineral was oligoclase from which recently sericitized, silicified xenomorphs aligned in streaks exist only. The phenoblasts of the younger plagioclase generation are fresh albites. The quartz shows a remarkable varying fabric. It consists of grains showing undulose extinction and forming mosaic-like contacts arranged into streaks and vein. The muscovite is the most frequent mineral of this rock-group. The primary and the younger generation could hardly be distinguished. Forming tress-like masses it separates itself from the salic components. Its young variations developed by transformation of biotite and felspar can frequently be observed. The primary biotite crystals are mostly decomposed to different extent. Most frequent phenomenon is the chloritization of the biotite crystals and/or their transformation into calcite-magnetite-muscovite. Small crystals of the newly formed biotite generation hardly occur in the mica-schist-like rocks. The garnet graine are decomposed in all of the cases. A number of transformation processes can be observed beginning from the chloritization along clefts to the garnet pseudomorphs filled by calcite and by chlorite.

Originally, the mica-schist-like rocks were mica-schist, gneisses composed of quartz, plagioclase, muscovite, biotite, garnet. The regional metamorphism of the source rock of dominantly sedimentary origin exceeded the degree of metamorphism in the greenschist facies. On the basis of the anortite content of the plagioclase (over 15%) and a widespread occurrence of the almandine garnet, these rocks could be ranged into the Barrow-type staurolite-almandine subfacies of the amphibolite facies. As a result of a younger regressive metamorphism, the primary mineral association has transformed and a secondary mineral assemblage consisting of

TABLE 1

Chemical compositions of the biotite-plagioclase gneiss occurrences

1. Mezőhegyes, Mezőkovácsháza, Végegyháza
2. Szank, Tázlár, Jánoshalma
3. Algyő, Ferencszállás
4. Endrőd, Kőrösszegapáti
5. Average chemical composition of the central biotite-plagioclase gneiss belt

	1			2			3			4			1—2	1—3	1—4	2—3	2—4	3—4	5
	$\overline{X}_{N=19}$	S_x	$S_{\bar{x}}$	$\overline{X}_{n=30}$	S_x	$S_{\bar{x}}$	$\overline{X}_{n=20}$	S_x	$S_{\bar{x}}$	$\overline{X}_{n=5}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,01}$	$t_{0,95=2,09}$	$t_{0,95=2,07}$	$t_{0,95=2,01}$	$t_{0,95=2,04}$	$t_{0,95=2,00}$	$\overline{X}_{n=74}$
SiO_2	63,65	3,65	0,84	62,54	3,39	0,62	61,48	7,05	1,58	62,73	3,05	1,36	−1,09	−1,20	−0,52	−0,72	−0,11	−0,36	62,55
TiO_2	0,60	0,21	0,05	0,59	0,21	0,04	0,54	0,23	0,05	0,65	0,26	0,12	−0,06	−0,17	−0,42	−0,16	−0,55	−0,49	0,59
Al_2O_3	17,21	1,15	0,26	16,41	1,42	0,26	16,12	1,56	0,35	15,07	1,67	0,75	+2,45	+2,45	+3,37	−0,83	−1,92	−1,34	16,45
Fe_2O_3	0,63	0,41	0,09	1,02	0,98	0,18	1,06	0,78	0,17	0,94	0,19	0,08	−1,54	−1,72	−1,67	−0,13	−0,18	−0,32	0,91
MnO	0,09	0,04	0,01	0,12	0,40	0,10	0,11	0,08	0,02	0,08	0,02	0,01	−1,67	−0,59	−0,83	−0,51	−1,99	−0,81	0,10
MgO	1,66	0,63	0,14	2,10	0,67	0,12	2,48	1,40	0,31	2,00	0,39	0,17	+2,55	+2,37	−1,18	−1,22	−0,39	−0,72	2,09
CaO	2,30	1,04	0,24	1,97	1,05	0,19	2,80	1,81	0,41	2,93	2,56	1,14	−1,11	−1,06	+2,25	+2,05	+3,05	−1,15	2,14
K_2O	2,59	0,50	0,11	3,34	0,92	0,17	2,12	0,90	0,20	1,84	1,07	0,48	+3,30	+2,10	+2,33	+4,78	+3,32	−0,47	2,71
H_2O	2,72	0,98	0,22	2,74	0,74	0,14	3,12	1,24	0,28	2,20	0,68	0,30	−0,05	−1,10	−1,13	−1,37	−1,51	−1,59	2,80
H_2O	0,18	0,09	0,02	0,17	0,13	0,02	0,17	0,25	0,05	0,16	0,10	0,04	−0,75	−0,47	−0,96	−0,36	−0,23	−0,10	0,17
P_2O_5	0,30	0,17	0,04	0,17	0,08	0,01	0,17	0,17	0,04	0,13	0,04	0,02	+4,63	+2,77	+3,10	−0,01	−1,10	−0,48	0,14
CO_2	1,27	1,26	0,29	1,69	1,70	0,31	1,51	1,49	0,33	3,42	1,72	0,77	−0,90	−0,86	+3,14	−0,36	+2,11	+2,49	1,65
Σ	99,60			99,84			99,89			99,51									99,51

TABLE 2

Chemical composition of the biotite-plagioclase gneisses

1. Pusztaföldvár
2. Algyő, Ferencszállás, Újszilvás
3. Ásotthalom, Kelebia
4. Chemical composition of the southern mica-schist range

	1			2			3			1—2	1—3	2—3	4
	$\bar{X}_{n=25}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=35}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=28}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,00}$	$t_{0,95=2,01}$	$t_{0,85=2,0}$	$\bar{X}_{n=88}$
SiO_2	54,23	6,85	1,37	56,56	6,29	1,06	54,78	6,29	1,19	−1,33	−0,36	−1,03	55,36
TiO_2	0,59	0,28	0,06	0,98	0,41	0,07	0,55	0,17	0,03	+4,13	−0,66	+5,17	0,73
Al_2O_3	20,64	6,05	1,19	20,85	4,13	0,70	22,40	3,63	0,69	−0,88	−0,51	−1,46	21,67
Fe_2O_3	0,91	0,71	1,14	1,51	0,69	0,12	1,20	0,61	0,12	+3,71	−1,95	−1,87	1,23
FeO	6,18	2,85	0,57	6,24	1,60	0,27	4,92	1,19	0,22	−0,18	+2,06	+3,64	5,79
MnO	0,16	0,08	0,02	0,21	0,32	0,05	0,10	0,05	0,01	−0,12	+3,45	+2,76	0,14
MgO	1,90	0,63	0,13	2,21	1,07	0,18	1,82	0,53	0,10	−1,34	−0,50	−1,78	2,00
CaO	0,82	0,56	0,11	1,30	1,02	0,17	0,77	0,43	0,08	+2,18	−0,26	+2,55	1,00
Na_2O	1,07	0,54	0,11	1,29	0,61	0,10	0,81	0,51	0,10	−1,42	−1,80	+3,28	1,07
K_2O	4,58	0,81	0,16	4,20	0,97	0,16	4,13	1,24	0,23	−1,67	−1,56	−0,20	4,28
$+H_2O$	3,77	0,92	0,18	3,73	0,97	0,16	4,35	0,78	0,15	−0,17	+2,42	−2,73	3,95
$-H_2O$	0,13	0,07	0,01	0,15	0,12	0,02	0,15	0,07	0,01	−0,20	−0,82	−0,44	0,14
P_2O_5	0,10	0,05	0,01	0,12	0,07	0,01	0,14	0,15	0,03	−1,31	−1,36	−0,87	0,12
CO_2	3,43	2,10	0,42	0,65	0,81	0,14	3,44	1,55	0,29	+6,89	−0,04	+9,01	2,34
Σ	98,51			100,00			99,56						99,82

albite-chlorite-calcite-muscovite-quartz, and belonging to the greenschist facies has developed.

Chemical analyses are available from the rocks of the southern mica-schist range only. According to the sampling frequency of the boreholes, mica-schist-like rocks of three different areas have been compared (Table 2).

The Al_2O_3 content (exceeding 20%), the K_2O/Na_2O ratio (heavly exceeding 1.0), the high FeO content refer to a sedimentary origin. The areal heterogenity of the source rock, the degree of the metamorphism, the selectiv effects of the secondary transformations have been studied by "t"-test. The author is of the opinion, that a significant difference could not be stated among the mica-schist locations. That is why a comprehensive mean is also given here.

Amphibolites

Amphibolites with a relatively limited areal extension and subparalell to the trend of the polymetamorphic formations have been uncovered at *Tázlár, Szank, Füzesgyarmat,* and — south of the zone above — at *Öttömös, Üllés, Algyő, Békéssámson, Battonya.* These amphibolites are compact, finely-crystallized, dark-green rocks of slightly schistose or non-schistose character. In addition to green-hornblende and plagioclase, epidote, quartz, titanite, rutile, apatite, garnet can be found as accessories.

The green hornblende crystals are mostly decomposed, transformed — in many cases into biotite. The majority of the amphibolites has supposedly been formed by metamorphism of very old ophiolites and bear traces of secondary transformations. There is not a significant difference among the chemical composition of the scattered amphibolite bodies (Table 3).

TABLE 3

Chemical composition of the biotite-plagioclase gneisses

1. Békésssámson, Battonya-E
2. Algyő
3. Szank
4. Üllés-NW, Öttömös

	1		2		3		4	
	$\bar{X}_{n=4}$	S_x	$\bar{X}_{n=4}$	S_x	$\bar{X}_{n=7}$	S_x	$\bar{X}_{n=4}$	S_x
SiO_2	47,82	1,33	45,35	3,21	49,32	2,16	50,90	2,08
TiO_2	1,07	0,35	1,37	0,76	2,16	0,78	1,56	0,45
Al_2O_3	16,67	2,82	15,10	0,69	15,67	1,56	14,88	2,20
Fe_2O_3	2,56	1,36	3,87	1,50	3,87	1,53	4,20	3,43
FeO	7,80	2,29	6,09	2,57	8,19	1,91	7,99	2,62
MnO	0,19	0,13	0,17	0,04	0,24	0,09	0,26	0,08
MgO	7,35	1,38	8,04	2,26	5,00	1,03	5,16	0,65
CaO	7,17	2,42	11,69	2,34	8,30	0,82	6,44	1,55
Na_2O	3,23	0,21	2,27	1,09	3,42	0,70	3,63	0,99
K_2O	0,51	0,27	0,59	0,76	0,85	0,20	1,14	0,49
$+H_2O$	3,59	1,46	2,64	1,77	2,05	0,48	2,61	1,55
$-H_2O$	1,19	1,31	0,04	0,04	0,31	0,21	1,01	1,80
P_2O_5	0,15	0,12	0,24	0,28	0,44	0,28	0,17	0,12
CO_2	0,51	0,17	2,82	4,81	0,14	0,20	0,32	0,30
Σ	99,81		100,28		99,96		100,27	

The rock types of the granitogenic ranges

Metatexites

The southeastern migmatite zone can be traced along a SW–NE trend at *Mezőhegyes, Dombegyháza, Mezőkovácsháza–SE, Kunágota.* The migmatite zone of Danube–Tisza Interfluve is less known. The boreholes finished recently uncovered the southern part of the migmatite zona in the environs of *Kiskunhalas,* while its northern zone became know at *Cegléd* and *Újszilvás.*

The rocks of the migmatite zones are metamorphites hit by a partial anatexis and in which the textural and mineralogical relicts and characteristics of the parent rock (=paleosome) are still identifiable, but — simultaneously — the new, re-melted part (=neosome) also appears. The neosome consists of the components of the leucosome. (The nomenclature of the migmatites and granitoids is used here according to MEHNERT [1968].)

The fabric of the metatexites is of lineation character. In the paleosome the original, oriented and schistose nature of the fabric has been preserved. The neosome frequently took the adventage of this inherited schistose, stromatic character of the paleosome and developed along the schistosity. The leucosome mobilizates are separated by melanosome streaks from the paleosome. The paleosomes in the Great Hungarian Plain are mostly biotite-plagioclase gneisses, rarely mica-schists. The leucosome mobilizates are composed of plagioclase, quartz, biotite. Their fabric is coarsely crystallized and slightly oriented. The most common components of the leucosome are subhedral plagioclases with a generally uniform optical appearance. The An-content of the plagioclase amounts to 15–20%. Another plagioclase generation also appears, showing a lower An-content and replacing the above mentioned plagioclase generation. Apart from the plagioclase, the other important mineral is the quartz. Being crystallized after the feldspars it occurs in form of xenomorphic crystals among those. Its recrystallization leads to replacement and resorption phenomena. The only mafic component is chloritized biotite.

The leucosome-paleosome contact is considered as a genetical feature. This contact shows an abrupt change in the mineralogical composition. In thin sections this contact is of more diffuse character. The amount of the biotite crystals in the leucosome increases towards the contact. The main component of the melanosome is the biotite. The melanosome bands consist of biotite, plagioclase, quartz and titanite crystals. The proportion of the biotite may amount to 40–50% of the rock part involved. The plagioclase is of 15—20% An-content, being in accordance with that of the plagioclase in the leucosome mobilizates is summarized in the Table 4.

Diatexites

In the southeastern part of the Great Hungarian Plain *(Battonya, Battonya–East, Dombegyháza–South, Kevermes, Sarkadkeresztúr)* and in some central parts of the Danube–Tisza Interfluve *(Soltvadkert, Soltvadkert–North, Nagykőrös, Kecskemét)* the crystalline basement complex is composed of granodioritic diatexites.

Being homogenized towards the granodioritic zone, the metatexites gradually pass into diatexites of granodioritic composition. Sometimes inclusion-like paleosome relicts still can be observed in the diatexites, the homogeneous rock, however, could not be devided into leucosome and paleosome anymore. The overwhelming majority of the rock mass has melted. The diatexites of the both zones mentioned above are granodiorites consisting of plagioclase, quartz, K-feldspar and biotite. The accessories are: apatite, zircon, epidote.

TABLE 4

The neosome differentiates of the migmatite range in the southeastern part of the Great Hungarian Plain

	1. Leucosome			2. Melanosome		
	$\bar{X}_{n=9}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=7}$	S_x	$S_{\bar{x}}$
SiO_2	69,14	4,26	1,42	50,08	5,03	5,90
TiO_2	0,27	0,18	0,06	0,96	0,43	0,16
Al_2O_3	16,75	1,89	0,63	15,85	2,25	0,85
Fe_2O_3	0,23	0,22	0,07	1,13	0,60	0,23
FeO	1,49	0,83	0,28	7,59	4,09	1,55
MnO	0,04	0,05	0,02	0,19	0,09	0,03
MgO	0,62	0,31	0,10	4,26	0,99	0,37
CaO	2,11	0,80	0,27	4,73	2,14	0,81
Na_2O	4,54	0,77	0,26	1,90	0,93	0,35
K_2O	2,41	0,92	0,31	3,89	1,42	0,54
$+H_2O$	1,26	0,40	0,13	3,15	1,20	0,45
$-H_2O$	0,12	0,05	0,02	0,85	0,74	0,28
P_2O_5	0,28	0,16	0,05	0,72	0,65	0,25
CO_2	0,52	0,65	0,22	4,68	4,27	1,62
Σ	99,78			99,98		

The hypidiomorphic plagioclase crystals are oligoclases of 15–20% An-content, being heavly sericitized, sometimes kaolinitized. The resorption of the oligoclase crystals at the rims is a phenomenon of common occurrence. At the rims of some oligoclase crystals fresh albite overgrowths developed being strikingly different from the sericitized inner part of the crystal. Besides this oligoclase generation composed of crystals with sericitized central part and albitic rims, appears a younger plagioclase generation characterized by blastic albite crystals of 2–7% An-content. Xenomorphic, hypidiomorphic crystals of this generation are generally fresh, sometimes slightly sericitized. The myrmekitic transformation has hit this plagioclase generation too. The young, acid plagioclase generation appears together with the blastic microcline, thus forming a joint association.

The diatexites contain two generation of K-feldspar. The older one — which had crystallized from melt and later was slightly transformed — is composed of microclines of medium triclinity. The younger generation consists of microclines with maximum triclinity, which developed in low-temperature and filled the intergranular space. It has been stated by optical measurements on universal stage [GY. BUDA, 1970] that the K-feldspars of the granodiorites in the Great Hungarian Plain show dual-triclinity similar to those in the granodiorites of the Mecsek Mts. The older generation is composed of intermedier microclines of medium triclinity, while the younger one does of microclines of maximum or almost maximum triclinity. The homogeneously distributed scales of the biotite are mostly chloritized.

The microfabric of the diatexites are characterized by a joint occurrence of several mineral associations by the abundant occurrence of replacement structures caused by the younger generations as well as by the presence of secondary mineral associations and mineral transformations caused by low-temperature metasomatism [Á. SZALAY, 1975].

The chemical composition of the two granodioritic zones shows a significant difference in the SiO_2, FeO, MgO, Na_2O content. The FeO, MgO proportion is

higher, the Na_2O/K_2O ratio is lower in the granodiorites of the Danube–Tisza Interfluve than those in the granodiorites of the southeastern part of the Great Hungarian Plain (Table 5).

TABLE 5

The chemical composition of the granodiorite ranges in the southeastern part of the Great Hungarian Plain and in the Danube—Tisza Interfluve

1. Battonya, Battonya-E, Mezőhegyes
2. Kecskemét, Nagykőrös

	1			2			1—2
	$\bar{X}_{n=17}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=3}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,10}$
SiO_2	68,08	1,00	0,24	65,45	1,80	1,04	+3,72
TiO_2	0,53	0,28	0,07	0,42	0,07	0,04	−0,58
Al_2O_3	15,90	0,77	0,19	15,55	0,22	0,12	−0,78
Fe_2O_3	0,61	0,20	0,05	0,75	0,23	0,13	−1,12
FeO	2,03	0,39	0,10	3,02	0,17	0,10	+4,23
MnO	0,10	0,05	0,01	0,04	0,01	0,01	−1,86
MgO	1,12	0,48	0,12	2,17	0,24	0,14	+3,87
CaO	1,76	0,46	0,11	1,97	0,47	0,27	−0,72
Na_2O	3,65	0,41	0,10	2,73	0,49	0,28	+3,51
K_2O	3,76	0,56	0,14	3,92	0,59	0,34	−0,48
$+H_2O$	1,61	0,31	0,07	1,96	0,37	0,21	−1,78
$-H_2O$	0,14	0,11	0,03	0,14	0,10	0,06	−0,01
P_2O_5	0,25	0,08	0,02	0,23	0,03	0,02	−0,32
CO_2	0,62	0,81	0,20	1,19	1,92	1,11	−0,95
Σ	100,16			99,54			

The diatexites are transversed by pegmatites. The anatectic rocks are interlaced by 10–50 cm thick, coarse-grained, pink pegmatite veins, consisting mostly of K-feldspar phenoblasts, quartz and muscovite. The pegmatites show a diffuse boundary zone without any mafic rim towards the country rock.

The pegmatites of the diatexites in the south-eastern part of the Great Hungarian Plain and in the Danube–Tisza Interfluve are characterized by the mean chemical compositions given in the Table 6. The chemism of the two pegmatite types is significantly different. The pegmatites in the Danube–Tisza Interfluve are more K-rich.

Crystalline schists hit by a single metamorphism

Mica-quartzites

Some boreholes have uncovered mica-quartzites in the *Ferencszállás* area and in the southern and southeastern part of *Ásotthalom* and *Pusztaföldvár* areas, respectively. These are pale-gray, schistose, frequently strongly folded, rigid rocks. Its dominant minerals are elongated quartz crystals with very undulose extinction. Besides muscovites and tiny biotites the rock contains a large quantity of sericite. These have developed by the decomposition of feldspars of sedimentary origin now being observed in form of hardly identifiable relicts. Rarely, fresh albite crystals can also be found in the rocks formed by the metamorphism of a psammitic sediment mass in the greenschist-facies. It is considered as an important genetic feature that this rock-type does not show any sign of polymetamorphism.

The mica-quartzites of quite different areas are of the same chemical composition and shows definite relationship in their composition (Table 7).

TABLE 6

The chemical composition of the pegmatites in the southeastern part of the Great Hungarian Plain and in the Danube—Tisza Interfluve

	1. SE Great Hung. Plain			2. Danube—Tisza Interfluve		
	$\bar{X}_{n=4}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=2}$	S_x	$S_{\bar{x}}$
SiO_2	63,12	2,75	1,38	64,99	0,18	0,13
TiO_2	0,14	0,07	0,03	0,02	0,01	0,00
Al_2O_3	17,49	1,40	0,70	14,77	0,18	0,13
Fe_2O_3	0,20	0,14	0,07	0,05	0,02	0,01
FeO	1,28	0,34	0,17	0,09	0,01	0,00
MnO	0,04	0,04	0,02	0,02	0,01	0,00
MgO	0,96	0,67	0,33	0,32	0,08	0,06
CaO	2,20	1,44	0,72	0,62	0,13	0,09
Na_2O	2,99	0,83	0,41	1,70	0,13	0,09
K_2O	8,47	1,71	0,86	13,52	0,38	0,27
$+H_2O$	0,95	0,28	0,14	0,41	0,01	0,01
$-H_2O$	0,10	0,03	0,01	0,05	0,03	0,02
P_2O_5	0,92	0,67	0,34	0,02	0,01	0,00
CO_2	1,02	0,25	0,13	—	—	—
Σ	99,88			96,58		

TABLE 7

The chemical averages of the mica-quartzites

1. Pusztaföldvár
2. Ferencszállás, Kiszombor
3. Ásotthalom

	1			2			3			1—2	1—3	2—3
	$\bar{X}_{n=9}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=6}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=6}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,16}$	$t_{0,95=2,16}$	$t_{0,95=2,22}$
SiO_2	71,68	3,38	1,13	72,70	0,83	0,34	72,78	3,06	1,25	−0,61	−0,64	−0,06
TiO_2	0,36	0,19	0,06	0,51	0,37	0,15	0,44	0,19	0,08	−1,05	−0,84	−0,40
Al_2O_3	13,57	2,18	0,73	14,51	1,93	0,79	12,27	2,00	0,82	−0,88	−1,20	−2,01
Fe_2O_3	0,50	0,25	0,08	1,26	1,36	0,55	0,91	1,15	0,47	−1,67	−1,04	−0,17
FeO	3,21	0,83	0,28	2,07	1,37	0,56	3,41	0,91	0,37	−2,02	−0,44	−2,00
MnO	0,11	0,02	0,01	0,04	0,03	0,01	0,10	0,04	0,02	+5,25	−0,80	+2,81
MgO	1,03	0,25	0,08	1,02	0,52	0,21	1,38	0,54	0,29	−0,32	−1,27	−1,18
CaO	0,67	0,28	0,09	0,32	0,26	0,11	0,58	0,14	0,06	−0,33	−0,73	−0,36
Na_2O	0,87	0,55	0,18	1,30	0,81	0,33	0,29	0,07	0,03	−1,21	+2,54	+2,99
K_2O	2,95	0,58	0,19	3,59	1,17	0,48	1,98	0,46	0,19	−1,42	+3,41	+3,22
$+H_2O$	2,27	1,04	0,35	2,23	0,79	0,32	2,55	0,70	0,29	−0,07	−0,09	−0,74
$-H_2O$	0,13	0,09	0,03	0,03	0,03	0,01	0,13	0,02	0,01	+2,62	−0,59	+6,64
P_2O_5	0,09	0,05	0,02	0,13	0,08	0,03	0,13	0,08	0,03	−0,37	−1,23	−0,59
CO_2	2,03	0,94	0,31	0,13	0,06	0,03	2,60	0,67	0,27	+4,88	−1,28	+9,03
Σ	99,47			99,84			99,52					

Chlorite schists, epidote gneisses

In the Tisza–Maros (rivers) junction (south part of the Great Hungarian Plain) unique rock-types were found in the basement complex of the Great Hungarian Plain. These two types could be genetically correlated.

The chlorite schists are greenish, fine-grained, compact schistose rocks. Their dominant mineralogical composition are: quartz, chlorite, albite, pyrite. Accessories are: calcite, muscovite, epidote. Their fabric is very characteristic one consisting of quartz grains and chlorite fibres aligned in one direction. The albite crystals link up with the quartz grains in an oriented way. The texture seems to be fresh the minerals belong to a single generation.

The quartz grains fitting together in a chain-like way form stripes of slightly undulose extinction. The plagioclase of albite composition is a fresh mineral being as clean as the water. The chlorites showing a greenish-brown colour under crossed nicols seem to be the dominant mafic component. Their fibrous appearance gives a strongly lineated character to the texture.

Inside the chlorite-schist sequence more coarse-grained rocks containing epidote and K-feldspar also appear. On the basis of their mineralogical composition and texture, these are considered as gneisses formed by K-metasomatism. The oriented character of their texture is less definite than that of the chlorite schists and becomes blastose. The effect of the blastesis manifests itself even in the chemical composition of the rock: the amount of the SiO_2 and K_2O has significantly increased than these of the chlorite schists (Table 8). The main mineralogical components of the epidote gneisses are as follow: quartz, plagioclase, epidote, muscovite, K-feldspar. The accessories are: pyrite, calcite, chlorite.

TABLE 8

The chemical composition of chlorite-schists and epidote-gneisses

1. Chlorite-schists (Algyő)
2. Epidote-gneisses (Algyő)

	1		2	
	$\bar{X}_{n=15}$	S_x	$\bar{X}_{n=13}$	S_x
SiO_2	60,59	5,07	71,61	3,52
TiO_2	0,07	0,10	0,11	0,11
Al_2O_3	15,13	1,98	13,74	1,93
Fe_2O_3	1,73	1,24	1,32	0,90
FeO	5,59	2,61	2,34	0,85
MnO	0,11	0,08	0,09	0,04
MgO	4,38	1,83	1,55	0,71
CaO	2,35	1,51	2,12	1,53
Na_2O	3,47	0,99	3,68	1,42
K_2O	0,50	0,29	1,23	0,64
$+H_2O$	4,01	1,45	2,13	0,48
$-H_2O$	0,11	0,07	0,06	0,05
P_2O_5	0,12	0,17	0,05	0,03
CO_2	1,45	1,65	0,25	0,34
Σ	99,61		100,28	

The mica-quartzites, chlorite-schists, epidote-gneisses and staurolite-mica-schists are products of a younger regional metamorphism in greenschist and/or amphibolite facies. These rocks were produced by a single metamorphism only. The epidote-gneisses bear marks of K-metasomatism.

Staurolite-mica schists

Staurolite-mica schists unknown structural position surrounded by granodiorites have been found in *Battonya-East* (SE-Hungary). These rocks are pale-gray, fine grained, slightly pressed metamorphites showing joints of schistosity with a silky shade. The lineated fabric of these rocks consists of isometric quartz grains as well as biotite and muscovite scales. The staurolite grains of 0.5–1.0 cm are scattered in the fabric of the rock. Accessories are as follow: magnetite, pyrite, zircon. Their chemical composition is characterized by a higher SiO_2 and CaO content as well as by a lower K_2O content than these in muscovite-biotite mica-schists (Table 9).

TABLE 9

The chemical composition of the staurolitic mica-schists discovered in Battonya-East

	$\overline{X}_{n=5}$	S_x
SiO_2	62,27	5,10
TiO_2	0,73	0,32
Al_2O_3	18,07	3,35
Fe_2O_3	1,30	1,04
FeO	3,81	0,80
MnO	0,16	0,06
MgO	2,14	1,08
CaO	1,60	1,10
Na_2O	1,14	0,90
K_2O	3,10	1,19
$+H_2O$	3,00	0,99
$-H_2O$	0,22	0,21
P_2O_5	0,08	0,05
CO_2	2,39	1,84
Σ	100,01	

Metasomatic rocks

Granites

The rocks of granitic chemical composition in the crystalline basement complex of the Great Hungarian Plain occur in forms of streaks and veins. In the diatexite-zones of granodioritic composition they are fine-grained, pink rocks with an aplitic appearance. Their fabric is fresh without any sign of secondary transformation. The rock is composed of quartz, K-feldspar, plagioclase and muscovite. The plagioclase is small, fresh albite. The K-feldspar is microcline of maximum triclinity. The aplites are of chemical composition being closest to that of granitic rocks (Table 10).

Medium-grained, pale gray granites have become know in the narrow belt from *Kiszombor* to *Kiskundorozsma* via *Ferencszállás–Algyő–Szeged*, at *Pálmonostora* as well as in the northern part of the *Szank* arch, and/or in the environs of *Endrőd*. Migmatite zone is not connected to these pale gray granites with slightly oriented fabric, wich are more coarse-grained than the aplites. In their environs the amount of the microcline-albite increases only. The rocks contain K-feldspar, quartz, plagioclase and muscovite. As accessories biotite, tourmaline, epidote and garnet can also be found.

TABLE 10

The aplitic rocks of the granodiorite range

1. Aplites of the Southeastern part of the Great Hungarian Plain
2. Aplites of the Danube—Tisza Interfluve

	1			2			1—2
	$\bar{X}_{n=17}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=3}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,10}$
SiO_2	72,75	1,64	0,40	73,20	1,13	0,65	−0,27
TiO_2	0,30	0,32	0,08	0,34	0,10	0,05	−0,21
Al_2O_3	14,49	1,02	0,25	14,34	0,93	0,54	−0,24
Fe_2O_3	0,29	0,18	0,04	1,04	0,46	0,27	+5,25
FeO	1,33	0,54	0,13	0,29	0,05	0,03	+3,25
MnO	0,09	0,05	0,01	0,03	0,04	0,02	−1,41
MgO	0,46	0,31	0,07	0,46	0,16	0,09	−0,01
CaO	1,02	0,42	0,10	0,58	0,10	0,06	−1,76
Na_2O	2,96	0,64	0,16	2,71	0,53	0,31	−0,63
K_2O	5,06	0,92	0,22	5,37	0,54	0,31	−0,56
$+H_2O$	0,95	0,38	0,09	1,41	0,18	0,11	+4,86
$-H_2O$	0,06	0,02	0,00	0,15	0,07	0,04	−1,99
P_2O_5	0,14	0,09	0,02	0,13	0,07	0,04	−0,20
CO_2	0,19	0,21	0,05	0,17	0,25	0,14	−0,13
Σ	100,09			100,22			

The microcline is a mineral of maximum triclinity wich has formed at a low temperature. Their crystals are fresh, products of blastesis and fill the intergranular space. They frequently contain graphic quartz and albite lamellae. The plagioclase is a mineral of two generations in this rock. The old plagioclase crystals are relicts of the source rock. The dominating plagioclase, however, is albite forming the younger generation. Big phenoblasts show a "chessboard-like" albite twinning. The microcline, quartz, albite, muscovite crystals belonging to the younger generation are of simultaneous origin and can be easily distinguished from the primary association composed of oligoclase, biotite and garnet.

The young granites are characterized by a high SiO_2 content and K_2O/Na_2O and/or K_2O/CaO ratios exceeding 3 (Table 11).

Albitites

The pale gray granites are accompanied by coarse-grained fresh albitites. The majority of the rockmass is composed of plagioclase crystals with 2–5% An-content showing "chessboard-like" twinning. The rocks contain quartz, K-feldspar and muscovite too. Apart from the young mineral association the transformed relicts of the source rock can also be observed.

In accordance with the mineralogical composition the Na is the dominant alkali. The SiO_2 content exceeds 70 w% (Table 12). The proportion of the TiO_2, MnO, Fe_2O_3, MgO is very low.

Albitic gneisses

Gneisses containing young mineral association are known in many areas, systematically in those wich are close to granites, albitites. Albeit the effect of developing new albite — microcline generation becomes already dominant, these

TABLE 11

The general composition of medium-grained granites

1. Deszk, Ferencszállás, Dorozsma
2. Szank, Pálmonostora, Jászszentlászló

	1			2			1—2
	$\bar{X}_{n=15}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=5}$	S_x	S_x	$t_{0,95=2,10}$
SiO_2	73,09	1,98	0,51	72,00	1,49	0,67	−1,12
TiO_2	0,15	0,25	0,07	0,27	0,23	0,10	−0,92
Al_2O_3	14,66	1,52	0,39	15,18	1,41	0,63	−0,61
Fe_2O_3	0,35	0,28	0,07	0,31	0,22	0,10	−0,38
FeO	1,37	0,61	0,16	1,40	0,64	0,29	−0,09
MnO	0,06	0,03	0,01	0,07	0,04	0,02	−0,19
MgO	0,41	0,40	0,10	0,42	0,23	0,10	−0,47
CaO	0,73	0,45	0,12	1,18	0,33	0,15	−2,01
Na_2O	2,85	0,51	0,13	3,34	0,54	0,24	+2,12
K_2O	5,10	1,13	0,29	4,75	0,36	0,16	−0,67
$+H_2O$	1,10	0,24	0,06	0,87	0,56	0,25	−1,35
$-H_2O$	0,05	0,04	0,01	0,24	0,14	0,06	+4,72
P_2O_5	0,13	0,09	0,02	0,05	0,02	0,01	+2,47
CO_2	0,08	0,10	0,03	0,13	0,20	0,09	−0,67
Σ	100,13			100,21			

TABLE 12

The chemical composition of the albitites discovered in the Ferencszállás—Deszk—Algyő area

	albitite		
	$\bar{X}_{n=3}$	S_x	$S_{\bar{x}}$
SiO_2	73,32	1,12	0,65
TiO_2	0,01	0,02	0,01
Al_2O_3	16,24	0,30	0,17
Fe_2O_3	0,56	0,24	0,14
FeO	0,86	0,23	0,13
MnO	0,09	0,05	0,03
MgO	0,16	0,14	0,08
CaO	0,76	0,31	0,18
Na_2O	4,84	1,50	0,87
K_2O	2,45	0,20	0,12
$+H_2O$	1,06	0,41	0,24
$-H_2O$	0,06	0,03	0,02
P_2O_5	0,23	0,04	0,02
CO_2	0,02	0,02	0,01
Σ	100,66		

metamorphites still contain identifiable relicts of the source rock. According to the author's observations made until recently the proportion of the albite generation is higher than that of the new feldspar generation, which is reflected in the chemical composition of the rocks as well (Table 13).

TABLE 13

The chemical composition of the albitic gneisses

1. Szank, Tázlár
2. Endrőd

	1			2			1—2
	$\bar{X}_{n=8}$	S_x	$S_{\bar{x}}$	$\bar{X}_{n=2}$	S_x	$S_{\bar{x}}$	$t_{0,95=2,30}$
SiO_2	67,40	5,62	1,99	71,92	1,88	1,34	−1,05
TiO_2	0,40	0,32	0,11	0,26	0,33	0,23	−0,54
Al_2O_3	14,40	0,98	0,35	14,27	0,78	0,55	−0,16
Fe_2O_3	0,82	0,76	0,27	0,31	0,11	0,08	−0,91
FeO	3,28	1,17	0,41	1,92	1,92	1,36	−1,33
MnO	0,12	0,03	0,01	0,03	0,01	0,01	+3,32
MgO	1,80	0,83	0,29	1,62	1,19	0,05	−0,26
CaO	2,15	0,79	0,28	1,45	0,47	0,33	−1,16
Na_2O	3,51	1,18	0,42	3,69	0,93	0,66	−0,19
K_2O	2,49	0,59	0,21	1,92	0,25	0,18	−1,28
$+H_2O$	1,50	0,42	0,15	1,81	0,39	0,28	−0,95
$-H_2O$	0,08	0,05	0,02	0,16	0,12	0,09	−1,63
P_2O_5	0,15	0,06	0,02	0,13	0,07	0,05	−0,27
CO_2	1,17	2,26	0,80	0,68	0,80	0,57	−0,29
Σ	99,27			100,17			

THE GENESIS OF THE METAMORPHIC AND GRANITOGENIC ROCKS

The overwhelming majority of the crystalline formations in the Great Hungarian Plain are *polymetamorphic* rocks. As a result of different metamorphic effects, which could be attached to different geotectonic cycles, the primary mineral association of the source rock has transformed and new associations have developed replacing the primary one. By distinguishing these the determination of the relative age and the metamorphic processes becomes possible in addition to drawing petrogenetic conclusions. The postulated process of the metamorphic evolution is outlined in *Fig. 2.* The qualitative changes of the metamorphic processes, the superimposing character of these concomitant transformation processes as well as the directions of the metamorphic transformation show a geological evolution.

The rocks infilling the one-time — probable Proterozoic — sedimantary basin were mostly sedimentary rocks, although a subordinate amount of magmatic rock was also present. *The effect of the first tectonometamorphic cycle was a regional one which caused a regional metamorphism of the rocks involved.* The sedimentary series being rich in pelitic components transformed into mica-schists the coarse-grained series greywackes did into biotite plagioclase gneisses. The scattered amphibolite bodies are probably the products of metamorphism of basic magmatism of initial character.

It is a hard job to evaluate the role of the Proterozoic plutonites. K. SZEPESHÁZY [1966] considers the crystalline basement of the Danube-Tisza Interfluve and that of the overwhelming majority of the Great Hungarian Plain as an ancient granitoid massif which has been hit by a regional metamorphism. The possibility could not be excluded that among the source rocks had been even granitogenic rocks, but the author is not in the position to show up any proof asserting this assumption.

The chemical composition and the uniform chemical character of the large central gneiss-range occupying a central position does not correspond to the granitic

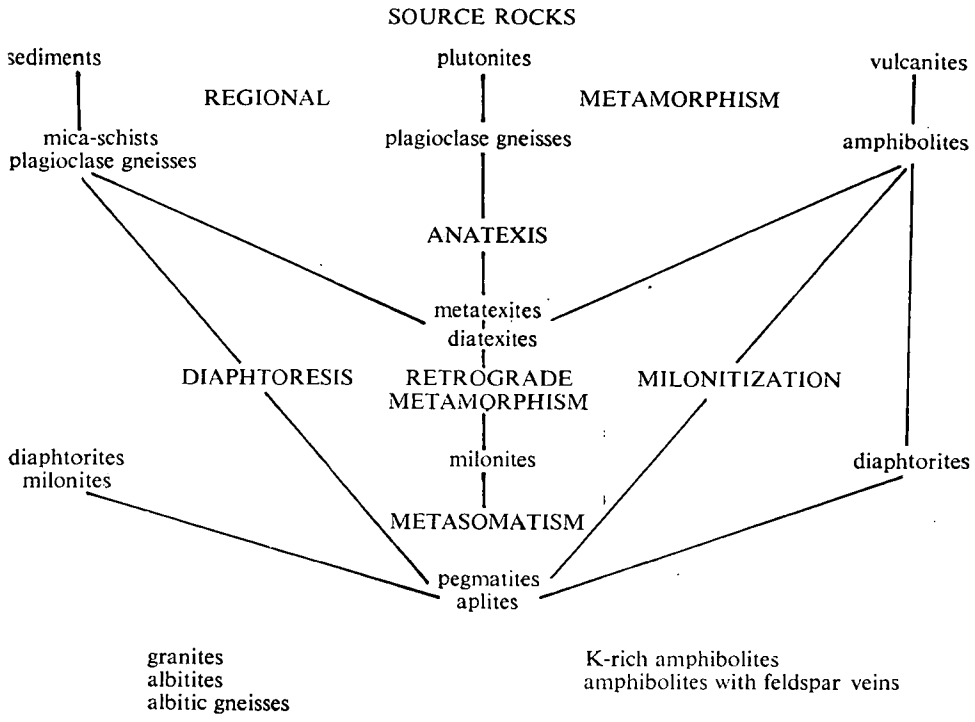

Fig. 2. Postulated process of the metamorphic evolution

or the granodioritic composition either. The 20–28% An-content of the primary plagioclases in the mica-schists and gneisses hints to the same i.e. sedimentary origin of the rocks. The lack of the primary feldspars do also assert the assumption that these rocks are mostly of sedimentary origin.

As a result of a regional metamorphism, they have formed in conditions equal to the Barrow-type staurolite-almandine or almandine-kyanite-muscovite subfacies of the amphibolite facies, and a primary quartz-plagioclase-biotite mineral association corresponding to the sedimentary composition has developed.

The polymetamorphic character of the rocks makes difficulties while studying the genetic relationship of the mica-schists and gneisses. The regional regularities could be elucidated by subsequent and more detailed mineralogical studies to be done, and by the spatial position of the index minerals.

The ultrametamorphic nature of the regional metamorphism locally and the onset as well as intensification of the anatectic process can be traced in the southeastern part of the Great Hungarian Plain and in the Danube–Tisza Interfluve. The neosome mobilizatum of the anatectites is a *venitic* product of a closed system differentiation produced by the partial melting of the source rock (paragneiss).

The geochemical analyses of the metatexites found in the Great Hungarian Plain have led to conclusions similar to those of K. R. MEHNERT [1968] concerning the genesis of the migmatites in the Black Forest (W-Germany) (Table 14). It can be stated from the distribution of the elements that closed system differentiation of the

TABLE 14

Comparison of the chemical composition of some migmatites, and their source rock from the southern part of the Great Hungarian Plain

	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	$+H_2O$	$-H_2O$
leucosome differentiate	69,14	0,27	16,75	0,23	1,49	0,04	0,62	2,11	4,54	2,41	1,26	0,12
melanosome differentiate	50,08	0,96	15,85	1,13	7,59	0,19	4,26	4,73	1,90	3,89	3,15	0,85
average of 3 leucosome and 1 melanosome differentiates	64,32	0,44	16,51	0,46	3,01	0,08	1,58	2,76	3,89	2,77	1,74	0,30
source rock	63,65	0,60	17,21	0,63	3,31	0,09	1,66	2,30	3,25	2,59	2,72	0,18
difference +	0,67							0,46	0,64	0,18		0,12
difference −		0,16	0,70	0,17	0,30	0,01	0,08				0,08	

paleosome has taken place (*i. e.* without accumulation of any material of significant amount from outside).

The most advanced stage of the anatexis is represented by the diatexites of granodioritic origin. In accordance with plagioclase crystals in the leucosome of the migmatite, its plagioclase crystals are oligoclases of 15–20% An-content. The primary K-feldspar making an appearance in the diatexites, however, suggests a more advanced stage of the melting as compared to the formation of the neosome in the migmatite. The partial melting of the K-feldspar-proof biotite-plagioclase gneisses resulted in metatexites, their full melting did in diatexites. The degree of melting of the source rock explains the absence of the K-felspars in the metatexites and/or their presence in the diatexites.

H. G. F. WINKLER [1967] has prooved by experiments, that at corresponding temperature a granodioritic melt may develop from K-felspar-proof, plagioclase-quartz-biotite bearing gneisses. The melt may contain different amount of K-felspar depending on the conditions of the melting. W. KNABE [1966] studied the anatexis of a material composed of biotite-quartz-plagioclase at a hydrostatic pressure of 2 Kbar. It has been stated that the biotite do not disappear totally at the beginning of the anatexis. A part of the biotite remains as resistite during the anatexis, while the other part yields K-feldspar components by the incongruent melting of the biotite.

At the starting phase of the anatexis a fragmentary part of the biotite transforms only, and that time a small amount of K-felspar develops as compared to the plagioclase, quartz. Concentrating the K-content the biotite separates itself in a form of resistites in the melanosome surrounding the leucosome. The latter developes in a lower temperature and concentrates the Si and Na content. Simultaneously to the increase of the temperature, however, the biotite also melts, the minerals cease to exist separately, a homogeneous melt developes which has a sufficient K-content for the crystallization of the primary feldspars.

The processes outlined here, i.e. the segregation, the melting and the subsequent K metasomatism which resulted in the development of the microcline-phenoblasts and pegmatites of the granodiorites, have been illustrated in triangle diagrams on the basis of chemical analyses *(Fig. 3)*.

The first metamorphic evolution cycle has finished with the palingenesis. The first tectonometamorphic evolution cycle resulted in the regional metamorphism of rock masses consisting of mostly pelitic sediments and greywackes in amphibolite facies. As a result of the regional metamorphism wide zones of mica schists gneisses, amphibolites have developed. Because of the steeper temperature gradient and the considerable amount of volatile components the rocks have partially or totally melted in the more heavly tectonized zones of the metamorphism, forming granodioritic rocks of palingenic origin.

In the contrary to the first one, the second metamorphic cycle has been a regressive one. The regressive metamorphism, milonitization of the rocks formed earlier have taken place. As a result of the regressive effects, mineral assemblages of the greenschist facies have developed in the metamorphites originally belonging to the amphibolite facies. The simultaneous nature of the regressive effect could be definitely stated by analysing the fabric, thus this process is considered as the result of a single geotectonic cycle. The regressive metamorphism may be allocated in time in between the regional metamorphism in the amphibolite facies and the younger regional metamorphism in the greenschist facies.

In the third significant tectonomorphic cycle the effect of the metasomatism is a decisive one. Locally and/or in one area even metamorphism in greenschist and/or amphibolite facies has taken place as a result of which chlorite schists, epidote-gneisses, mica-quarzite and/or staurolitic mica-schist have developed.

The regional metamorphism is characterized by low-temperature mineral associations of similar age. The young mineral association occurs in rock-forming amount in the aplites, medium-grained granites, albitites, albitic gneisses, while in the whole metamorphic complex manifests itself in form of blastic transformation marks.

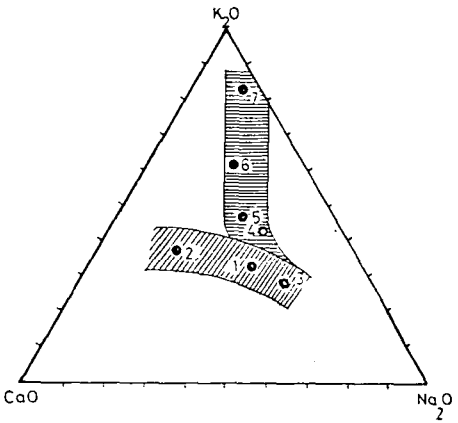

Fig. 3. Anatectic and metasomatic processes of the crystalline rocks in the Great Hungarian Plain.
1. Biotite-plagioclase gneiss of SE part of the Great Hungarian Plain
2. melanosome (SE part of the GHP)
3. leucosome (SE part of the GHP)
4. granodiorite (SE part of the GHP)
5. granodiorite (Danube—Tisza Interfluve)
6. pegmatite (SE part of the GHP)
7. pegmatite (Danube—Tisza Interfluve)

During the metasomatic process of regional extent the Si, K and Na were remobilized *(Fig. 4)*. A part of the mobilizates causing the metasomatic transformation originated from the source rock itself. Their migration has taken place within a short distance and local realignement of the materials has happened. All the alkalis, however, could not be stemmed from the rocks involved. It may be postulated that the water released in supracritical condition could remobilize during its migration the alkali content even of the deeplying rocks. Becoming spatially differentiated during their migration, the Na and K have formed granites rich in microclines and young albitites rich in plagioclase in the upper zones and along the tectonic lines.

The granitoids of other origin are the results of the evolutional phases of the process of the orogenesis. *Synkinematic* and *latekinematic* granitoides could equally be found in the areas discussed here. (Classifying the granitoids the MARMO's classification principles based on ESKOLA's magmatectonic system have been applied.)

Synkinematic rocks of the migmatite-granodiorite ranges surrounding the central gneiss-mica schist range. The characteristics of the synkinematic granitoids are as follow: source rock of dominantly sedimentary origin, anatectic formation and a presence of a wide migmatite zone. Their chemical composition is granodio-

ritic; the K_2O/CaO ratio exceeds 1. (This ratio 3.76/1.76 and 3.92/1.97 in the cases of the granodiorites of the Great Hungarian Plain and that of the Danube–Tisza Interfluve, respectively.) The granitizing effect has hit the synkinematic granodiorites later. The only K-felspar component is the microcline which is mostly younger

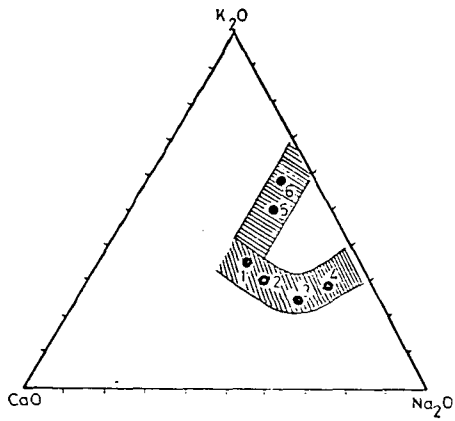

Fig. 4. Post-crystallizational Na and K metasomatism of the crystalline rocks in the Great Hungarian Plain.

1. Biotite-plagioclase gneiss (average of the GHP)
2. Albitic-gneiss (Szank, Tázlár)
3. Albitic gneiss (Endrőd)
4. Albitite (Ferencszállás, Deszk)
5. Granite (Ferencszállás, Deszk, Kiskundorozsma)
6. Granite (Szank, Pálmonostora, Jászszentlászló)

than the primary mineral association and it is considered as a result of a younger K-mobilizing geotectonic cycle.

The late-kinematic granites are represented by younger medium-grained granites, found in forms of narrow zones within the ancient crystalline rocks. Their chemical composition is granitic, the K_2O/CaO ratio systematically exceeds 2 (in aplites 5.06/1.02 and/or 5.37/0.58, in the medium-grained granites 5.20/0.33 and/or 4.75/1.18).

The narrow, belt-like form and the lack of migmatization in the country rock refer to *the tectonically defined character of the late-kinematic granite formation.* On the basis of their mineral associations of the same age (*i.e.* microcline with high triclinity and plagioclase of albitic composition) *the late-kinematic granites are as metasomatic formations.*

Postkinematic granites have not uncovered until recently in the area involved. The author is of the opinion that the ortoclase bearing granites of the granitoid zone south of the Lake Balaton represent this group.

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MINERALOGICAL AND CHEMICAL STUDIES ON ANTHOPHYLLITE-ACTINOLITE SCHIST FROM WADI UM KABU, SOUTH EASTERN DESERT, EGYPT

A. A. EL SOKKARY

ABSTRACT

The present work deals with the mineralogy and chemistry of an amphibole schist from Wadi Um Kabu lying in the south Eastern Desert of Egypt. The detailed mineralogical investigation including optical and X-ray diffraction studies revealed this schist to be composed of anthophyllite, actinolite, goethite and calcite. Chemical analysis showed the studied schist to be enriched in MgO content (28.79%), but contains certain amounts of FeO (5.29%), CaO (5.42%) and Al_2O_3 (3.57%). It is concluded that the original monoclinic actinolite was transformed to orthorhombic anthophyllite during a thermal metamorphic pulse and in an atmosphere of CO_2 and H_2O, while the exuded Ca and Fe formed the two minerals calcite and goethite.

Thus the schists of Wadi Um Kabu were the place of differential mobilization of certain elements particularly Ca and Fe during thermal metamorphism. This mostly happened under amphibolite facies of metamorphism. The studied amphibole schist sheet lying between muscovite-talc schists can be envisaged as an original basic intrusive body that was subjected later to metamorphism giving rise to the present amphibolite rock.

INTRODUCTION

Wadi Um Kabu is an eastern tributary of the larger and well known Wadi Gemal lying in the southern parts of the Eastern Desert of Egypt *(Fig. 1)*. Wadi Um Kabu consists of a series of diorite gneisses beside schists of various types, the most important of which is the muscovite-talc schist. Sometimes these schists are associated with the so-called actinolite schist variety, the subject of the present study. For details of the field occurrence of these gneisses and schists, see EL SOKKARY [1960].

Other actinolite schist occurrences are reported in literature though not studied in detail. BASTA and ZAKI [1961] described an actinolite-biotite schist from Wadi Sikait area in the south Eastern Desert. EL SHAZLY and HASSAN [1972] mentioned the presence of actinolite bands associated with talc schists in Wadi Sikait–Wadi El Gemal area.

The Um Kabu actinolite schist is subjected here to detailed mineralogical and chemical studies. The mineralogical work includes megascopic description of the hand specimens, specific gravity determination, optical characters under the microscope, X-ray diffraction and chemical mineral tests. A complete chemical analysis of the actinolite schist is given as compared with other analyses quoted from literature. On this basis a discussion of the possible genesis and metamorphic history of the particular type of schist under study is given.

Admittedly, the various types of schists of the basement rocks of Egypt are among the least studied rocks particularly from mineralogical and chemical points

of view. The present work is a contribution to the mineralogy and chemistry of certain members of these schistose formations occurring within the basement rocks of Egypt.

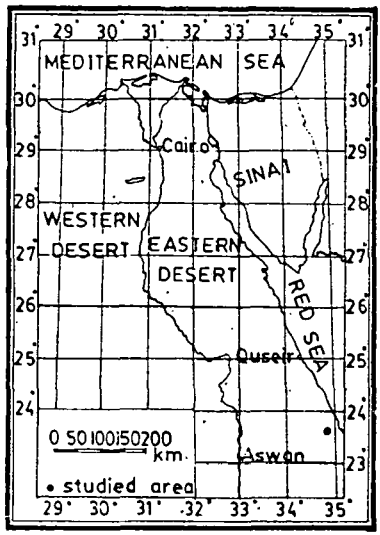

Fig. 1. Map showing the location of Wadi Um Kabu area

MINERALOGICAL INVESTIGATIONS

The field term actinolite schist refers to a rock that is composed of aggregates of long prismatic needle-shaped crystals which may be greater than 3 cm in length, with acicular habit and pale greenish color. Associated with some yellowish brown ferruginous material, mostly goethite, which may be an alteration product of the green amphibole. Some specimens develop minor muscovite and talc components. Average specific gravity on three hand specimens is found to be 3.06. Specimens of almost pure actinolite schist are chosen for more detailed studies.

Under the microscope, the amphibole shows prismatic crystals ranging in color from pale to dark greenish shades, possibly reflecting variable quantities of iron, weakly pleochroic from pale greenish to somewhat dark greenish but certain varieties may show relatively stronger absorption than others, with two perfect sets of cleavage and the presence of cross fractures. Statistical measurement of extinction angle on 50 grains revealed that about 50% of them show straight extinction while 50% give extinction angle ranging from 10–24°, crystals are sometimes strained but the phenomenon is not strong. Interference colors are yellow of the second order indicating moderate birefringence. Occasionally with tiny opaque or transparent inclusions.

The fact that with respect to the extinction angle, the grains fall into two groups of almost equal proportions: a group with straight extinction angle and the other with oblique angles ranging from 10–24°, indicates that the amphibole schist under study may be composed of a mixture of two varieties: one probably anthophyllite and the other is actinolite. Few grains of yellowish mineral occasionally with anomalous birefringence are observed, they mostly represent goethite flakes and constitute a minor fraction of the total grains.

A sample of the amphibole schist is powdered to pass 150 mesh size. It is subjected to X-ray diffraction using a Philips chart recorder diffractometer adapted to work with Cu radiation, wavelength 1.5418 Å, Ni filter, current 26 mA, voltage 36 kV and scale 16.64. Table 1 gives the partial diffraction pattern of the sample as compared with corresponding data for standard (ASTM) anthophyllite, actinolite and goethite.

TABLE 1

Partial diffraction pattern of the amphibole schist sample as compared with standard anthophyllite, actinolite and goethite

Sample		Anthophyllite[1]		Actinolite[2]		Goethite[3]	
d Å	I/I_0	*d* Å	I/I_0	*d* Å	I/I_0	*d* Å	I/I_0
9.41	88	9.30	25				
9.12	10	8.90	30	9.06	40		
8.42	88	8.26	55	8.42	70		
7.14	14	7.48	7				
5.10	3	5.04	13	5.12	30		
4.75	8	4.90	9	4.88	50		
4.60	13	4.62	13				
4.53	5	4.50	25	4.52	50		
4.21	22	4.13	20			4.21	100
3.88	4	3.90	13	3.87	50		
3.55	9	3.65	35				
3.39	1	3.36	30	3.38	70		
3.28	10	3.24	60	3.27	60		
3.12	100	3.05	100	3.11	80		
2.95	2	2.87	20	2.94	60		
2.81	24	2.84	40				
2.71	5	2.74	20	2.71	100		
2.70	2	2.68	30			2.69	80
2.60	2	2.59	30	2.59	50		
2.54	2	2.54	40	2.53	60		
2.45	1	2.43	13			2.44	70
2.38	2						
2.34	2	2.32	20	2.33	40		
2.32	2	2.29	20	2.27	20		
2.29	1	2.25	13				
2.17	2	2.17	9	2.16	40		
2.12	1	2.14	30				
2.02	1	2.07	9	2.04	30		
2.01	2	2.06	9	2.01	30		
2.00	2	1.99	15				
1.89	5	1.88	11				
1.87	3	1.84	20	1.86	20		
1.81	2	1.73	30				
1.65	2	1.69	13	1.65	40		
1.64	1	1.64	9				
1.62	3	1.62	30				
1.61	2						
1.59	1	1.58	20	1.58	40		
1.56	1						
1.44	2			1.43	40		

1: ASTM Card No. 9—455.
2: ASTM Card No. 7—336.
3: ASTM Card No. 8—97.

It is concluded from Table 1 that the amphibole schist sample is essentially composed of the following main minerals: anthophyllite, actinolite and goethite. On the basis of the diffraction pattern alone, it looks that anthophyllite is more abundant followed by actinolite while goethite is a minor constituent. However, it is to be noted that the *d* spacings of the ASTM anthophyllite are not exactly matching the corresponding lines of the investigated sample. This may be attributed to certain chemical differences between the sample anthophyllite and the ASTM anthophyllite, a matter which will affect though to a limited extent the dimensions of the unit cell of each mineral.

Two chemical mineral tests are done in order to check the presence of goethite and any traces of carbonates. In the first experiment the sample powder is boiled in HCl (1:1) for about one hour, the filtrate is tested by potassium ferrocyanide when the Prussian blue color characteristic for Fe is obtained. Thus acid leachable iron is present in the form of an independent mineral that is soluble in HCl and takes the ferric state, this mineral is mostly goethite. In the second experiment, the sample powder is tested by adding few drops of HCl and observing the reaction by a hand lens. Weak effervescence is observed from few grains. Traces of carbonates possibly as calcite are therefore present associating the amphiboles.

To sum up, these mineralogical investigations revealed that the pure amphibole schist under study is composed of: anthophyllite, actinolite, goethite and traces of carbonates as calcite. The latter two components, namely goethite and calcite, may represent alteration products of the actinolite.

CHEMICAL ANALYSIS

Table 2 exposes the complete chemical analysis in weight per cent of the oxides of the amphibole schist sample compared with the corresponding analyses of anthophyllite and actinolite as given by DEER *et al.* [1972].

TABLE 2

Chemical analysis (wt.% of the oxides) of the amphibole schist sample compared with other analyses of anthophyllite and actinolite

	Amphibole schist	Anthophyllite	Actinolite
SiO_2	51.35	58.48	51.40
Al_2O_3	3.57	0.57	3.88
Fe_2O_3	0.29	0.58	3.90
FeO	5.29	7.85	14.91
MnO	0.50	0.27	0.33
MgO	28.79	29.25	11.22
CaO	5.42	0.14	10.17
Na_2O	0.34	0.08	1.67
K_2O	0.18	0.02	0.09
TiO_2	0.01	0.03	0.74
P_2O_5	0.05	—*	—*
H_2O^+	3.44	2.60	1.90
H_2O^-	0.21	0.20	0.04
Total	99.44	100.07	100.25

*: Not reported.

As it is clear from Table 2, the most characteristic feature of this amphibole schist is its enrichment in MgO content that equals 28.79%. Nevertheless, the amphibole is not precisely an anthophyllite on account of the presence of relatively considerable amount of CaO that equals 5.42%. The substitutions of FeO (5.29%) and CaO (5.42%) for MgO in the analysed amphibole are not quite extensive (causing no pronounced decrease in the MgO content) to produce a true ferroactinolite. There is also moderate substitution of Al_2O_3 mostly in place of SiO_2 that reaches 3.57%. It is to be noted that the Fe_2O_3 content (0.29%) of the amphibole schist sample is too low particularly if it is taken into consideration that the sample contains free goethite phase visible by the naked eye in hand specimens.

The above chemical considerations indicate clearly that the amphibole schist sample under study is composed neither of anthophyllite alone nor actinolite alone, rather it is composed of a mixture of the two minerals with more anthophyllite due to enrichment in MgO in accordance with the results of X-ray diffraction.

GENETIC DISCUSSION

Results of microscopic investigation together with X-ray diffraction analysis and chemical analysis revealed that the amphibole schist under study is composed mineralogically of anthophyllite, actinolite, goethite and calcite. Anthophyllite may be more abundant than actinolite. The two minor minerals goethite and calcite are alteration products of the actinolite.

It seems that the monoclinic calcic amphibole actinolite has undergone a transformation reaction during a thermal event by means of which it lost part of its Fe and Ca (causing relative enrichment in MgO) and converted to orthorhombic anthophyllite. The extra Fe and Ca are exuded and formed the two minerals goethite and calcite, respectively. This transformation happened during a thermal metamorphic event and in an atmosphere of H_2O and CO_2. On the basis of this study, it is possible to say that the present anthophyllite-actinolite schist was subjected at least two metamorphic cycles: one that produced the original actinolite schist and the other that caused partial transformation of actinolite to anthophyllite.

The above paragenetic sequence receives the following evidence from the field [EL SOKKARY, 1960]. The talc-muscovite schists interveined by the actinolite schist of Um Kabu contain sporadic and abundant crystals of ferriferous calcite that attains very big sizes. The latter mineral can be considered to form small scattered pockets within the host schists. This attests that the ferriferous calcite is later than the actinolite schist and is formed during a metasomatic process accompanying a thermal event.

It appears that the schists of Wadi Um Kabu were the place of certain chemical mobility under thermal metamorphic environments. The main elements that were mostly affected by differential mobilization in the present case are Ca and Fe. If however the mentioned ferriferous calcite proved to be Mg-bearing, it would be possible to say that the element Mg has suffered as well from limited mobility. This case of differential mobility of elements has resulted in a sort of metamorphic differentiation of the original amphibole schist to give amphibole-calcite-goethite association.

FAIRBRIDGE [1972] mentioned that the process of metamorphic differentiation is essentially one of redistribution of elements: Ti, Fe, Mg and Ca segregated from Si, Al, and Na with little change of bulk composition. Differential solubility under conditions of heterogeneous pressure in foliated rocks plays an important role.

The amphibole schist sheet lying between muscovite-talc schists can be visualized as an original basic intrusive body taking the form of a sill that was subjected later to metamorphism giving rise to the present amphibolite rock or anthophyllite-actinolite schist.

The grade of metamorphism of this amphibole schist rock lies within the amphibolite facies. TURNER and VERHOOGEN [1960] pointed out to the cummingtonite (or anthophyllite)-tremolite assemblage (the closest to the present anthophyllite-actinolite assemblage) to belong to the almandine-amphibolite facies. WINKLER [1967] stated that orthorhombic amphiboles i.e. anthophyllite and the Al-bearing gedrite are formed at the beginning of the hornblende-hornfels facies. Since anthophyllite is an important constituent of the present anthophyllite-actinolite assemblage, therefore this assemblage is mostly formed in the hornblende-hornfels facies or the amphibolite facies.

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NEW CONODONTS FROM THE NORTHHUNGARIAN TRIASSIC

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ABSTRACT

In this paper three new conodont species are described from the eastern end of the Alsóhegy Karstplateau (southernmost unit of the Silica-nappe) and from the Rudabánya Mountains: *Hindeodella (Metaprioniodus) longobardica* n. sp., *Gondolella auriformis* n. sp., *Metapolygnathus baloghi* n. sp. Finally, some amplifications is added to the knowledge of the *Gondolella foliata* (Budurov) and *Prioniodina tatrica* (Zawidzka). The material described here is reposited in the collection of the Geologic and Paleontologic Department of the József Attila University, Szeged.

Hindeodella (Metaprioniodus) longobardica n. sp.

Pl. I, *Figs. 1–3*, Pl. II, *Fig. 2.*

1972 *Hindeodella (Metaprioniodus)* n. sp. A — Kozur & Mostler, p. 18, Taf. 14, *Fig. 14*

Derivatio nominis: from the occurence in the Longobardian.
Holotypus: the specimen No. 1, Pl. I, *Fig. 3,* Pl. II, *Fig. 2*
Stratum typicum: Longobardian Nádaska Limestone.
Locus typicus: the eastern end of the Alsóhegy Karstplateau, Silica nappe, North Hungary.

Fig. 1. Position of the Alsóhegy (1) and the Rudabányaian-Mts. (2) in North Hungary

Diagnosis: Bars are about of the same length. The first or rarely the second denticle of the anterior bar and the last or next to the last denticle of the posterior bar are longer; sometimes may almost reach the length of the main cusp.

Description: The unit is straight or slightly laterally curved and gently arched. The bars are about of the same long and both bear 3–6 denticles. The first or rarely the second denticle of the anterior bar and the last or next to the last one of the posterior bar are longer; sometimes almost as long as the main cusp. The remain denticles of the anterior bar are somewhat smaller than those of the posterior bar.

The main cusp is of central or subcentral position. All denticles except the larger one of the anterior bar incline slightly posteriorly.

The basal groove is narrow but relatively deep, extends the entire length of the basal edge and enlarges into a compressed but deep cavity beneath the main cusp.

Remarks: This new species stands nearest to the *Hindeodella (Metaprioniodus) multihamata* (HUCKRIEDE), from which it differs by the considerably smaller growth and by that fact that the first denticle is the largest on the anterior bar, before which only rarely can be found a small one.

Occurrence: Up to now this new species has been known from the Longobardian of the Balaton Highland and the eastern end of the Alsóhegy Karstplateau (both Hungary); on the later place constitutes a characteristic Longobardian conodont-assemblage together with *Metapolygnathus mungoensis* (DIEBEL), *Gondolella foliata* (BUDUROV) and *Prioniodina tatrica* (ZAWIDZKA). This species may probably be a guide form of the Longobardian substage.

Material: 65 specimens.

Gondolella auriformis n. sp.

Pl. I, *Figs. 4, 5;* Pl. II, *Fig. 1;*
Pl. III, *Fig. 1;* Pl. VIII, *Fig. 1*

Derivatio nominis: latin, *auris,* meaning ear; referring to the in oral view ear-like platform.

Holotypus: the specimen No. 1, Pl. III, fig. 1.

Stratum typicum: Upper Longobardian red limestone.

Locus typicus: on the ridge south from the north-western side-valley No. 8 of the Telekes valley, Rudabányian Mountains, North Hungary.

Diagnosis: Anterior carina high, fused, posteriorly rapidly decreases in height. Platform short, but wide; in oral view ear-like. Pit is of terminal position on the keel.

Description: The carina is composed of 6–9, rarely 10–11 denticles. Its anterior part high, fused; posteriorly rapidly decreases in height. The main cusp is stronger; in our material there are only three specimens having a small denticle behind it.

The platform begins between the centre and the posterior third of the unit. Its anterior or near to the centre part is the widest, then tapers rapidly; giving together with the upturned platform-margins the the ear-like shape of the platform in oral view. The platform-end is rounded or sometimes squared off, but with rounded corners. On some extreme forms a narrow platform-rudiment can be seen (Pl. I, *Fig. 4),* which may extend to the anterior third of the unit. Near to the platform-end a constriction may appear on one or both side, without taxonomic value. The microstructure of the platform is prismatic (Pl. VIII, *Fig. 1).*

The keel is wide with groove and ends terminally beneath the main cusp in a flaring pit.

Remarks: The new species is in transitional position between the genera *Gondolella* and *Metapolygnathus.* Because species with high carina *(G. excelsa)* and with short platform *(G. tadpole)* can be found in the former genera, as well, and this species has a terminal pit, it is placed still into the genera *Gondolella.*

Gondolella auriformis developed probably from the *Gondolella excelsa* (MOSHER); and bound by a transitional series with the *Metapolygnathus baloghi* n. sp. Therefore, forms can be found within this species, on which the platform-margins on one or both sides are drawn into a node (Pl. I, *Fig. 4*). The boundary between the two

species has been established by that fact, that those specimens having at least on one side two marginal nodes belong already to the *Metapolygnathus baloghi* n. sp.

The *Gondolella auriformis* n. sp. differs from the *Metapolygnathus parvus* KOZUR by the terminal position of the pit and the wider, ear-shaped platform; from the *Gondolella tadpole* HAYASHI by the higher and shorter carina and the non-thickened platform-margins.

Occurrence: ?Middle—Upper-Longobardian; it has been known only from the Rudabánya Mountains up to now.

Material: 68 specimens.

Metapolygnathus baloghi n. sp.

Pl. III, *Fig. 2;* Pl. IV, *Fig. 1;*
Pl. V, *Figs. 1, 2;* Pl. VII. *Figs. 1, 2*

1975 *Epigondolella carnica* sp. n. — KRYSTYN, in KRISTAN-TOLLMANN—KRYSTYN, p. 273—275, only the specimen on Taf. 3, *Fig. 4*

Derivatio nominis: in honour of Prof. Dr. K. BALOGH.

Holotypus: the specimen No. 1, Pl. IV, *Fig. 1*

Stratum typicum: Upper Longobardian red limestone.

Locus typicus: on the ridge south from the north-western side-valley No. 8 of the Telekes valley, Rudabányaian Mountains, North Hungary.

Diagnosis: Very wide, posteriorly tapering, platform, which extends the posterior third—half of the unit. It is widest between its anterior third—half, here with nodes. Anterior carina high, fused, posteriorly rapidly decreases. Pit terminal.

Description: The carina is composed of 7–11 denticles. Its anterior part high, fused; posteriorly rapidly decreases in height. The last denticle, as main cusp, is stronger.

The platform begins between the centre and the posterior third of the unit. On its widest part, which is between the anterior third—half of it, there are 1–4 nodes on both sides, but at least 2 nodes on one side. In our material some specimens can be found with less wide platform and more upturned platform-margins: probably they represent the transition to the *Metapolygnathus carnicus* (KRYSTYN). Posteriorly the platform rapidly tapers; the platform-end rounded or sometimes squared off, but with rounded corners. No nodes are on this part. The microstructure of the platform is prismatic. (Pl. VII, *Figs. 1, 2*).

The keel is wide, with groove, ends terminally beneath the main cusp in a flaring pit.

Remarks: The *Metapolygnathus baloghi* n. sp. differs from any other species of the genera *Metapolygnathus* by its very wide, posteriorly rapidly tapering platform. The relation with the *Gondolella auriformis* n. sp. was discussed above.

Occurrence: Uppermost Ladinian in the Rudabánya Mountains; Lower Carnian at Saklibeli, Taurus Mts., Turkey.

In the type locality from the sample No. R—67 the following conodonts have been recovered, which are interesting of our point of view:

Metapolygnathus baloghi n. sp.	40 exemplars
Gondolella auriformis n. sp.	67 exemplars
Gondolella foliata (BUDUROV)	19 exemplars
Gondolella polygnathiformis BUDUROV & STEFANOV	4 exemplars
From the sample No. R—66, one metre below the R—67:	
Gondolella foliata (BUDUROV)	105 exemplars
Gondolella auriformis n. sp.	1 exemplars

As it can be seen, the first representatives of the *G. polygnathiformis*, beside the predominance of the *G. foliata*, have already appeared in the sample No. R—67. Because they take their first appearance in the *archelaus*zone at Saklibeli (KRYSTYN, 1975), we assume theUpper Longobardian age of this conodont assemblage.

Material: 40 specimens.

Gondolella foliata (BUDUROV, 1975)

Pl. VI, *Figs. 2, 3*

1975 *Paragondolella foliata* sp. n. — BUDUROV, p. 79—80, Taf. 1, *Fig. 1—22*
1975 *Gondolella excelsa* (MOSHER) — KRYSTYN, in KRISTAN-TOLLMANN—KRYSTYN, Taf. 3, *Fig. 7*
1976 *Paragondolella foliata* BUDUROV — BUDUROV, p. 101, Taf. II, *Fig. 18, 35*

Remarks: In our material (more than 250 specimens) all transition can be found between forms with rounded platform-end and loop and forms with squared off platform-end and loop (such as the holotype). Therefore the latter feature seems to be an intraspecific variation.

The *Gondolella foliata* represents the transition between *Gondolella excelsa* (MOSHER) and *Gondolella polygnathiformis* BUDUROV & STEFANOV. At the eastern end of the Alsóhegy the last representatives of *G. excelsa* occur in the *hungaricus*-subzone and *G. foliata* appear first in the *mungoensis*-zone. Already MOSHER [1973, p. 150] reported from this transition between *G. excelsa* and *G. polygnathiformis*, however, at that time the species *G. foliata* was not yet established.

In the investigated area this species ranges at least to the base of the range of the *G. polygnathiformis*.

Prioniodina tatrica (ZAWIDZKA, 1972)

Pl. VI, *Fig. 1;* Pl. VII, *Fig. 2*

1972 *Neospathodus tatricus* sp. n. — ZAWIDZKA, p. 462—463, *Fig. 2*, Pl. 1, *Figs. 1—3, 5*
1974 *Prioniodina tatrica* (ZAWIDZKA) — BECHSTÄDT—MOSTLER, p. 40—41.

Remarks: In the investigated area this species does not occur in the fairly rich Pelsonian–Fassanian conodont-fauna; but frequent in the Longobardian. Only one specimen was found at the base of the range of the *Gondolella polygnathiformis* BUD. & STEF.

Other occurrences: Upper Anisian? or Lower Ladinian? of the Choč nappe, Tatra Mts., Poland; Longobardian of the Northern Limestone Alps.

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EXPLANATION OF THE PLATES I—VIII

PLATE I

1. *Hindeodella (Metaprioniodus) longobardica* n. sp. Spec. No. 3. T—364*. 200x
2. *Hindeodella (Metaprioniodus) longobardica* n. sp. Spec. No. 2. T—364. 200x
3. *Hindeodella (Metaprioniodus) longobardica* n. sp. Holotypus. T—364. 3a:200x; 3b: The basal edge with the keel and the groove. 325x
4. *Gondolella auriformis* n. sp. Spec. No. 3. Transitional form to the *Metapolygnathus baloghi* n. sp., the platform-margins are drawn into a node on both side. R—67. 360x
5. *Gondolella auriformis* n. sp. Spec. No. 7. Juvenile form. R—67. 270x

PLATE II

1. *Gondolella auriformis* n. sp. Spec. No. 2. R—67. 1a: 360x; 1b: 270x
2. *Hindeodella (Metaprioniodus) longobardica* n. sp. Holotypus. T—364. The basal cavity beneath the main cusp. 970x

PLATE III

1. *Gondolella auriformis* n. sp. Holotypus. R—67. 360x
2. *Metapolygnathus baloghi* n. sp. Spec. No. 2. R—67. 270x

PLATE IV

1. *Metapolygnathus baloghi* n. sp. Holotypus. R—67. 270x

PLATE V

1. *Metapolygnathus baloghi* n. sp. Spec. No. 8. R—67. 270x
2. *Metapolygnathus baloghi* n. sp. Spec. No. 6. Juvenile form. R—67. 270x

PLATE VI

1. *Prioniodina tatrica* (ZAWIDZKA). T—364. 270x
2. *Gondolella foliata* (BUDUROV). T—364. 140x
3. *Gondolella foliata* (BUDUROV). R—67. 140x
4. *Gondolella polygnathiformis* BUDUROV & STEFANOV. R—67. 140x

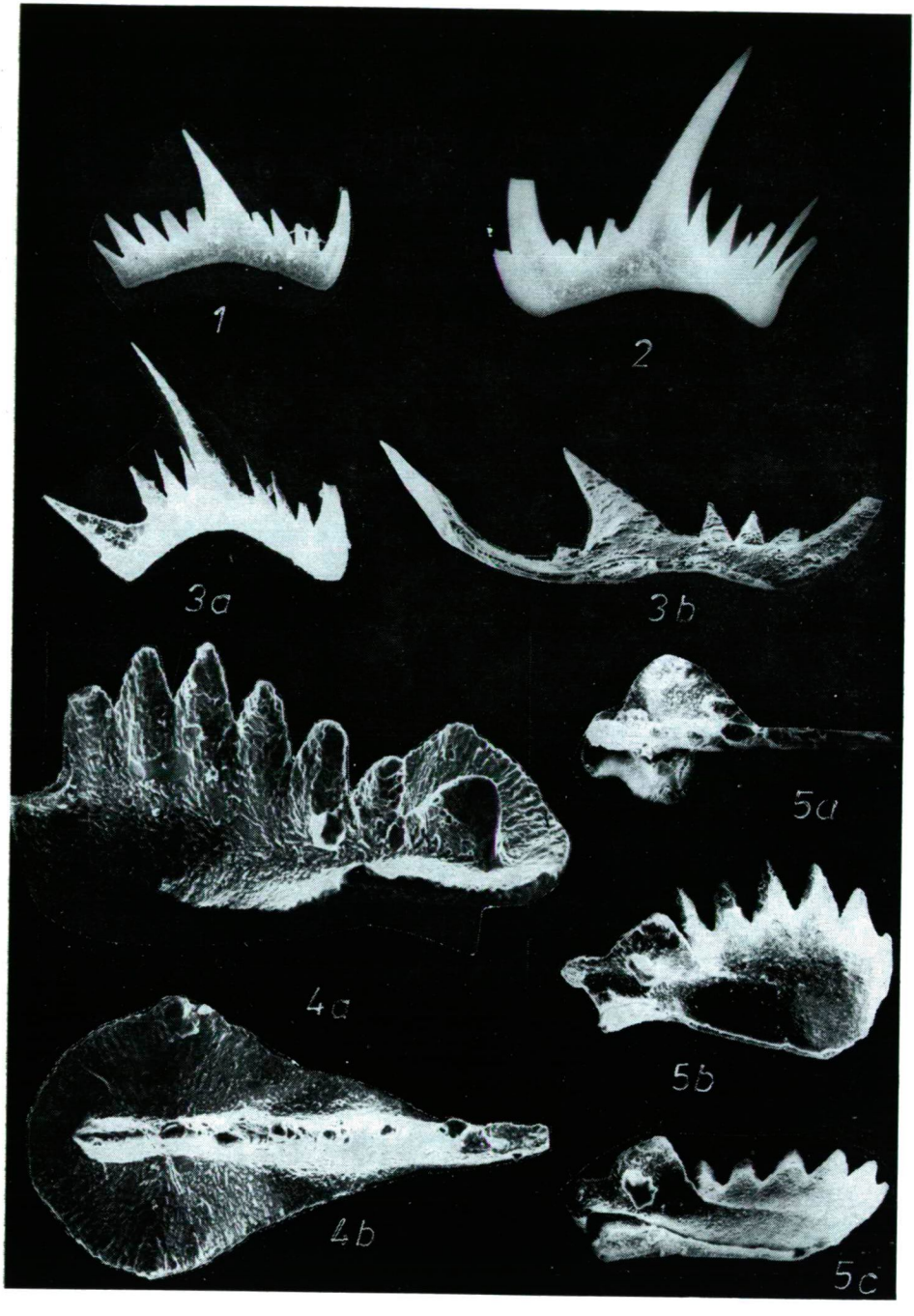
PLATE VII

1—2. *Metapolygnathus baloghi* n. sp. Spec. No. 2. 650x
   1. The prismatic microstructure of the platform.
   2. The aboral view of the platform with the flared pit.

PLATE VIII

1. *Gondolella auriformis* n. sp. Spec. No. 3. The prismatic microstructure of the platform. Oral view. 1300x
2. *Prioniodina tatrica* (ZAWIDZKA). The striated surface of the main cusp. 790x

* Samples: T—364: from the eastern end of the Alsóhegy-Karstplateau; R—67: from the Rudabányaian-Mts.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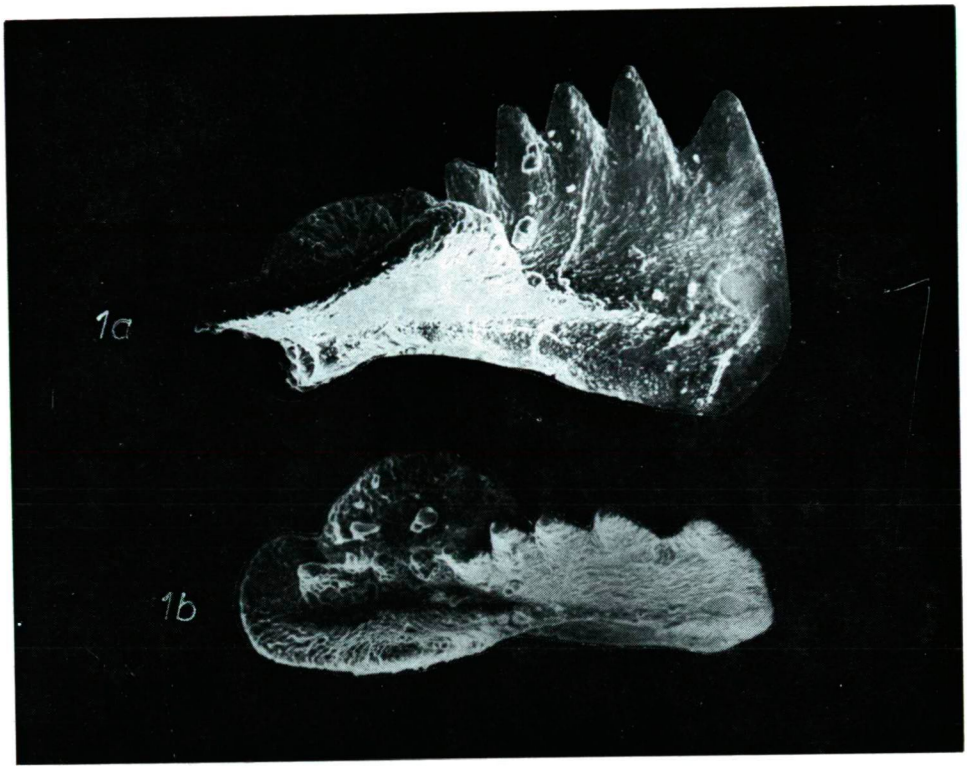
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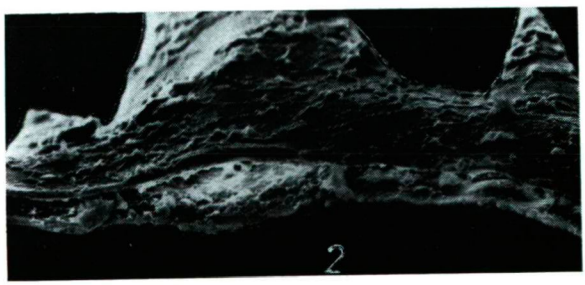
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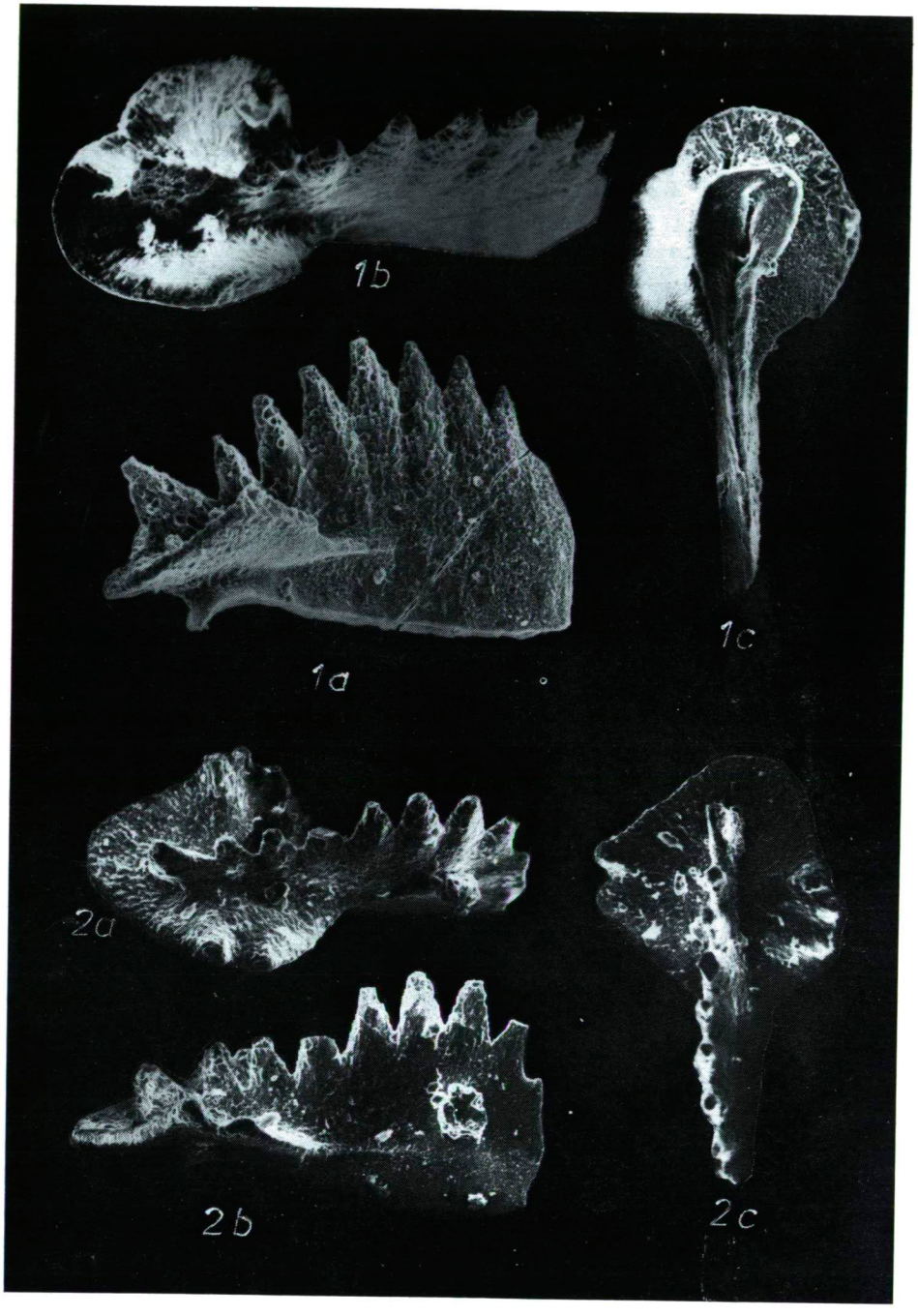
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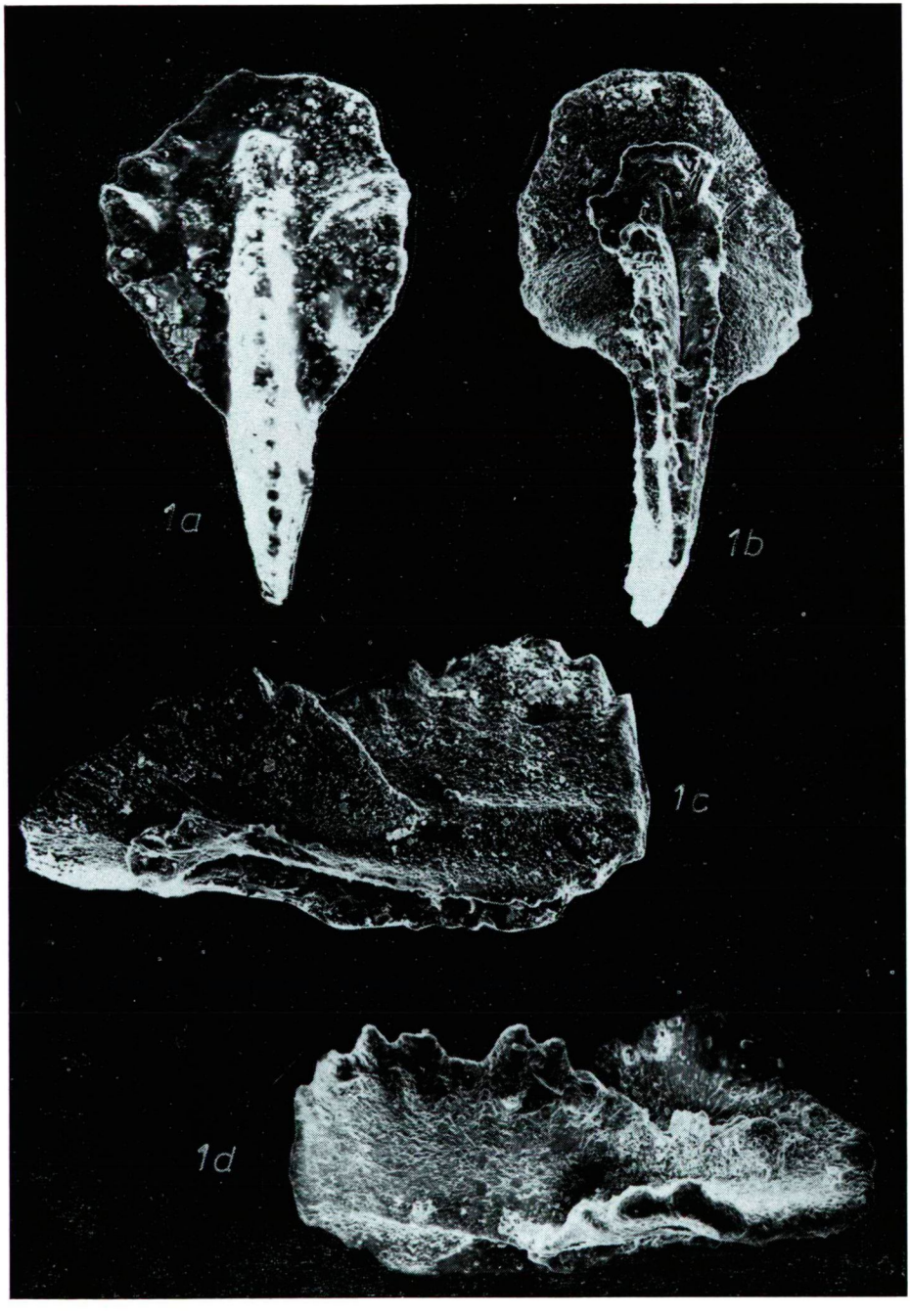
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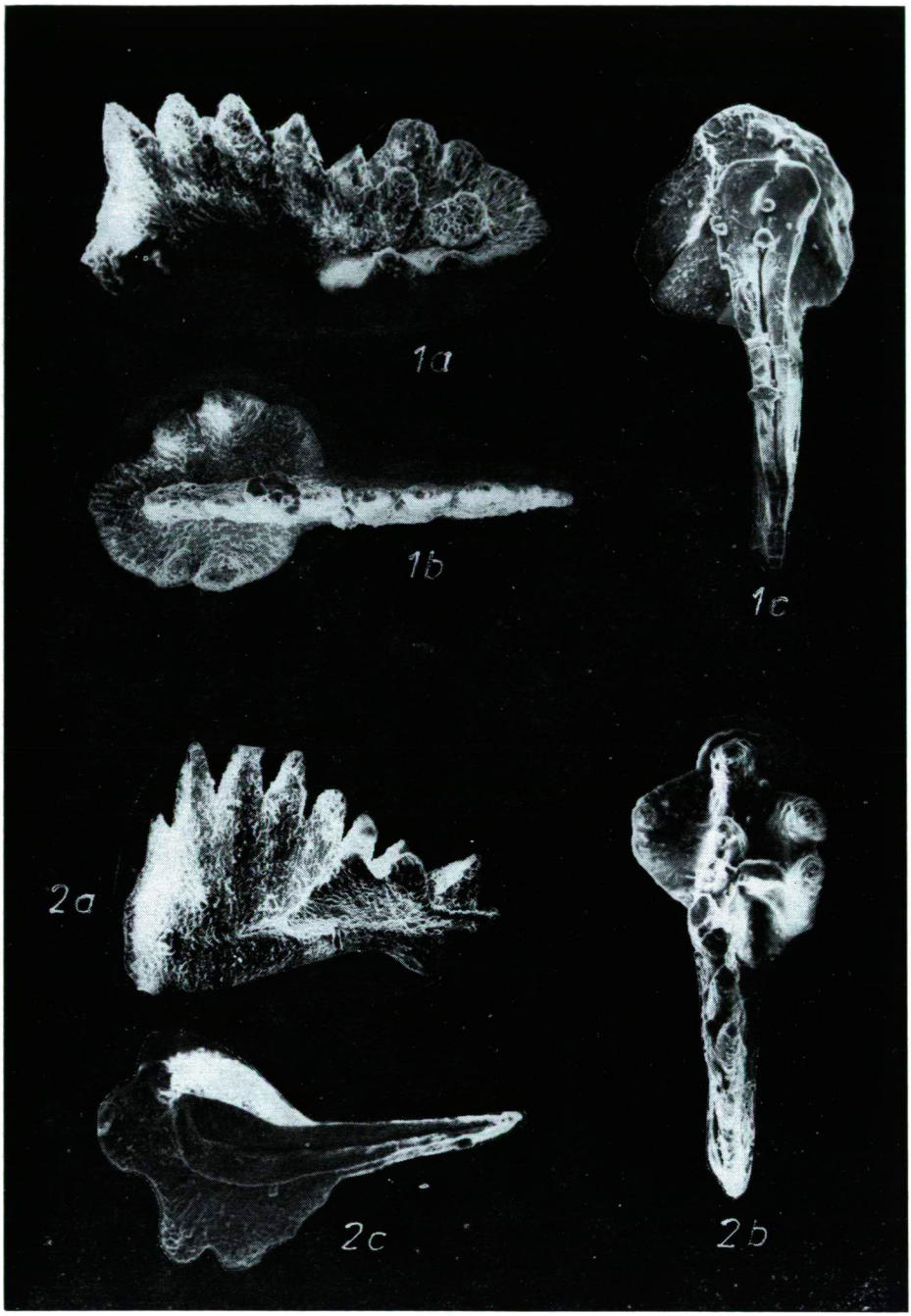
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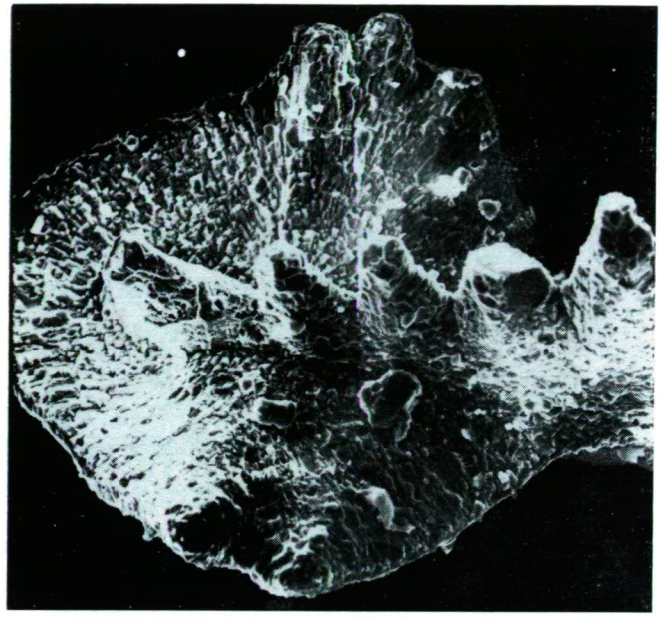

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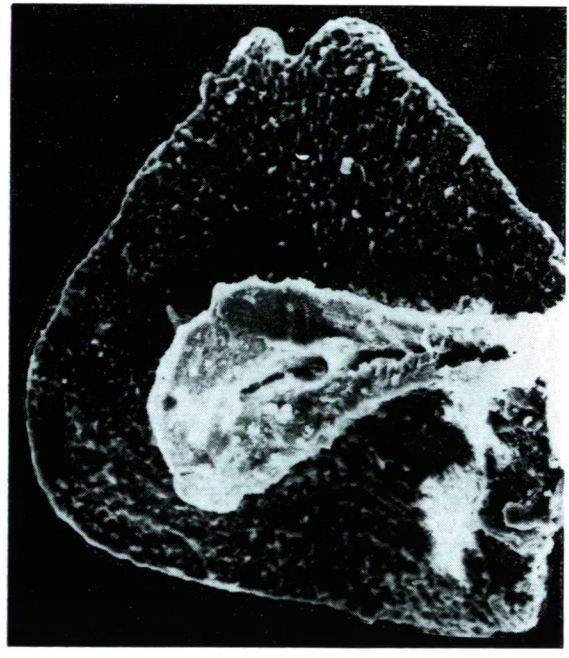

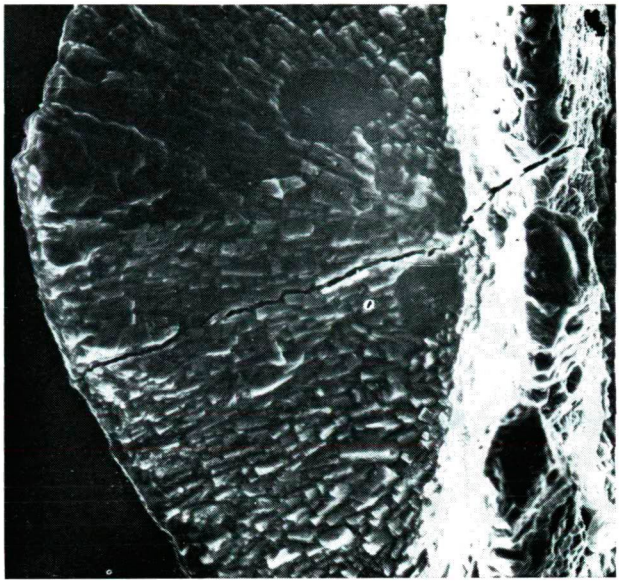

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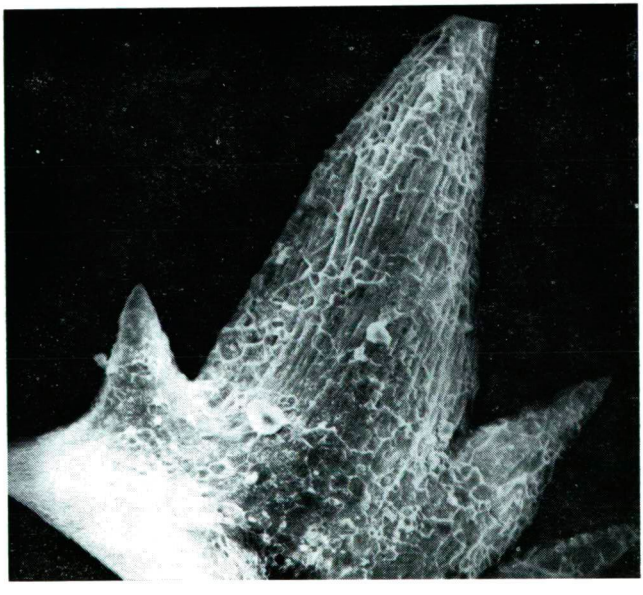

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ON THE AGE OF THE PALEOZOIC OF THE UPPONY MOUNTAINS (NORTH HUNGARY)

H. KOZUR and R. MOCK

SUMMARY

The Paleozoic sequence of the Uppony mountains begins with a *massive limestone complex* of some hundreds meter thickness. The age is assumed to be Middle Devonian from the geological position below the Upper Devonian. A 1200—1400 m (according to K. BALOGH, 1964) thick sequence of shales with intercalations of mostly thin-bedded limestones (in the lower part frequent, in the upper parts more rare), diabases and tuffitic layers *(limestone—shale complex)* follows above the massive limestone complex. By the aid of conodonts the age of this limestone—shale complex could be determined to range from Upper Devonian up to the lower part Middle Carboniferous (Bashkirian). Above this complex the more than 900 m thick *sandstone—shale complex* with some lydites and manganese oxide, but without any limestones, follows. From the geological position immediately above a thin limestone layer with Bashkirian conodonts this complex should be Middle Carboniferous and it seems therefore to be the immediate base of the sandstone—shale complex of Bashkirian to Lower Moscovian age in the Bükk mountains. The upper part of the limestone—shale complex with only minor amounts of limestones and the sandstone—shale complex are regarded as equivalents of the "Hochwipfel Beds" of the Southern Alps as of similar sequence in the Dinaric Medvednica Mountains. The stage of preservation of the conodonts in the Uppony Paleozoic is the same as in the Triassic of the Bükk mountains and Meliata-series as well as of parts of the Rudabánya Triassic. There are no differences in the metamorphosis between the Middle Devonian to Middle Carboniferous of the Uppony, the Lower Carboniferous of Ochtiná (Gemeric Palaeozoic) and the Triassic of Bükk mountains and Meliata series. Therefore an Alpine low grade metamorphosis of the Middle Devonian to Triassic beds is assumed in the studied area. The Hercynian orogeny seems to have little or no significance in the Uppony-Bükk area. No Hercynian metamorphosis or angular discoformities occur from the Middle Devonian to the Permian.

1. SHORT HISTORIC REVIEW OF THE RESEARCHES CARRIED OUT IN THE PALEOZOIC OF THE UPPONY MOUNTAINS

J. BÖCKH (1867) divided the Uppony Mountains into two formations: *1.* the light massive limestone was regarded by him as Jurassic, and *2.* the shale complex with limestone, lydite and sandstone intercalations as Upper Carboniferous age. — The massive Uppony Limestone was assigned by Z. SCHRÉTER (1945) to the Devonian? *(Series No. 1);* the overlying schists and limestones to the Lower Carboniferous *(Series No. 2)* and the upper part of the profile (schists and sandsones) to the Lower to Upper Carboniferous *(Series No. 3).* — G. PANTÓ (1954) distinguished also three complexes but all of them assigned to the Lower Carboniferous. — According to Á. JÁMBOR (1961) the Paleozoic of the Uppony Mountains should be older than the Moscovian verified in the Bükk Mountains (? Lower Carboniferous). This classification is based first of all on the fact that the facies of the *Series No. 3* is similar to that of the Kulm-formation. — K. BALOGH (1964) regarded the Paleozoic of Uppony Mountains as cover of the Paleozoic from Szendrő. According to him the massive limestone in the north *(Series No. 1)* was assigned to the Tournaisian, the higher

members *(Series No. 2 and 3)* to the Visean. He assumed a Sudetian phase between the Paleozoic of the Uppony Mountains and the Upper Carboniferous of the Bükk Mountains. Similarly to Á. JÁMBOR (1961) he also stated that in the Szendrő Mountains the Paleozoic should be stronger metamorphized. However, macrofossils (especially corals) have been found only in the Szendrő Mountains. K. BALOGH and L. KŐRÖSSY (1974, p. 393, Table 2; classification after J. ORAVECZ) suggested the "Lower Series" to begin both in the Szendrő and in the Uppony Mountains in the Ordovician. The 2nd and 3rd series are assigned by them to the Silurian in the Uppony and to the Silurian and Devonian in the Szendrő Mountains.

The lithological description of the individual sequences of the Uppony Paleozoic can be found in the book of K. BALOGH, 1964.

2. FOSSIL CONTENT OF THE INVESTIGATED SAMPLES

11 samples were investigated from the limestone—schist sequence and from the massive limestone of the Uppony Mountains within the scope of the studies concerning the stratigraphic subdivision of the Gemeride Paleozoic and the clearing its tectonic position in the Alpine-Carpathian region *(Fig. 1)*.

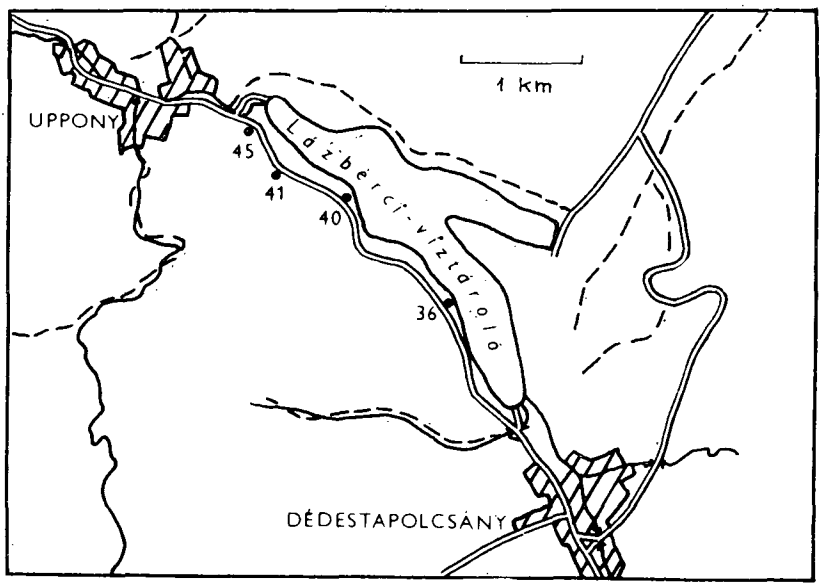


Fig. 1. Conodont-sampling points along the road between Dédestapolcsány—Uppony.

The first Paleozoic conodonts of Hungary have been found in four samples and these allowed the exact arrangement of the limestone—schist complex of the Uppony Mountains. The foregoing investigations on conodonts proved to be negative here (M. HAJÓS, 1971 and H. W. FLÜGEL 1975), however, this is caused undoubtedly not by the overshadowing effect of epimetamorphic processes but rather by the primary scarcity of conodonts in most of the Paleozoic strata of the West-Carpathians.

Sample 36

Idiognathoides sinuatus HARRIS et HOLLINGSWORTH, 1933, left and right forms (three specimens).
? *Idiognathoides noduliferus inaequalis* HIGGINS, 1975 (one fragment).

Sample 45 (Fig. 2)

Dryphenotus bilineatus (ROUNDY, 1926) (two well identifiable specimens; the *Dryphenotus* fragments occurring in a great number and in which the platform is broken, belong for the most part to this species).
Dryphenotus semiglaber (BISCHOFF, 1957) (one juvenile specimen).
Polygnathus sp., probably *P. inornatus* BRANSON, 1934 (one fragment).

Sample 40 (Fig. 3)

Bispathodus aculeatus (BRANSON et MEHL, 1934) (one specimen).
Bispathodus bispathodus ZIEGLER, SANDBERG et AUSTIN, 1974 (frequent).
Bispathodus costatus (BRANSON et MEHL, 1934) (very frequent).
Bispathodus cf. *spinulicostatus* (BRANSON, 1934) (one specimen).
Bispathodus ultimus (BISCHOFF, 1957) (frequent).
Palmatolepis gracilis gracilis BRANSON et MEHL, 1934 (frequent).
Palmatolepis gracilis sigmoidalis ZIEGLER, 1962 (rare).
Polygnathus communis BRANSON, 1934 (rare).
Polygnathus cf. *inornatus* BRANSON, 1934 (= *Polygnathus* n. sp. *A* GROESSENS, 1975, in sense of R. DREESEN, M. DUSAR and E. GROESSENS, 1976), (four specimens).
Polygnathus sp. (rare).
Pseudopolygnathus trigonicus ZIEGLER, 1962 (two specimens).
Pseudopolygnathus sp. (three specimens).
Numerous compound conodonts as well as *Spathognathus* sp.

Sample 41

Nothognathella sp. (one specimen).
Palmatolepis minuta minuta BRANSON et MEHL, 1934 (two specimens).
Palmatolepis gracilis gracilis BRANSON et MEHL, 1934 (one specimen).
Polygnathus cf. *lauriformis* DREESEN et DUSAR, 1974 (three specimens).
Different compound conodonts.

Notes on taxonomy: One of the specimens of *Palmatolepis minuta* is preserved in its total length, but its platform edges are corroded. Thus, the form seems to be slender than it is really. On the basis of the hardly curved blade this form can be compared only with *P. minuta minuta*. Another fragment belongs probably also to this species.

As it has been stated by R. DREESEN, M. DUSAR and E. GROESSENS (1976) among many *Bispathodus* species there are so uninterrupted transitions, that the validity of some species is ambiguous. This is true first of all in the cases, if the transitional forms are not bound to a definite stratigraphic horizon where the transition from one species into an other can be step-by-step followed. Between the species of the *bispathodus* and *aculeatus* line there are also transitional forms, in accordance with the statement of R. DREESEN, M. DUSAR and E. GROESSENS. Obviously, the two lines cannot be so clearly separated, as it often seems to be permormable. The existence of the morphotypes 1 and 2 of *B. costatus* evidences this very clearly. Both in this and all in similar cases it seems justifiable to perform no species separation, if the ranges concur completely or approximately with another and the transitional forms are available from the most different horizons. This is the case e.g. at *B. ultimus* and *B. ziegleri*. In the sample 41 much more transitional forms appear

between these two "species" than definite end-members of the uninterrupted transitional set which could be called either *B. ultimus* or *B. ziegleri*. In the transition forms the basal cavity reaches up to or nearly up to the posterior end as in the case of *B. ultimus;* it is however very narrow and more or less rudimentary behind the wide front part corresponding to the pit of *B. ziegleri*. The secondary set of denticles begins only behind the wide part of the basal cavity (similarly to the *B. ziegleri*). Both the length and width of the basal cavity and the beginning of the secondary set of denticles change very strongly and independently of each other. Since both species have the same stratigraphic range (see W. ZIEGLER, CH. A. SANDBERG and R. L. AUSTIN, 1974) the *B. ziegleri* (RHODES and AUSTIN-DRUCE, 1969) is regarded as younger synonym of the *B. ultimus* (BISCHOFF, 1957). Both "species" can be considered only as morphological end-members of a variation line consisting of numerous transitional forms. Numerous transitional forms exist also between *B. costatus* morphotype 1 and *B. bispathodus,* as it has been noted by R. DREESEN, M. DUSAR and E. GROESSENS (1976). However since both species have different stratigraphic ranges according to W. ZIEGLER, CH. A. SANDBERG and R. L. AUSTIN (1974 a, b), they will be treated here as separated species in spite of the fact that in consequence of the great number of transitional forms a separation in the available material seems to be very arbitrary. The transitional forms between *B. costatus* and *B. ultimus,* appearing similarly but only very sporadically, will be regarded as such transitional forms that can be nearly always observed at the transition of a species into another within the stratigraphic sequence.

According to H. KOZUR and H. MOSTLER, 1976 the *Gnathodus mosquensis* PANDER, 1856 (type species of *Gnathodus* PANDER, 1856) derived from the Upper Carboniferous (in sense of the Soviet classification: Stephanian) belongs to *Streptognathodus* STAUFFER et PLUMMER, 1932, wich is so the younger synonym of *Gnathodus* PANDER, 1856. Independently of that, if the concept of H. KOZUR and H. MOSTLER, 1976 is followed (i.e. *Streptognathodus* is a younger synonym of *Gnathodus*) or the opinion of S. ALEKSEEV (Conference on the Carboniferous, Moscow, 1975) is accepted (i.e. *Gnathodus* PANDER is restricted to *Gnathodus mosquensis* and it is regarded as nomen dubium) the term *Gnathodus* cannot be applied longer for the Lower and Upper Carboniferous conodonts being assigned till now to this genus. The genus *Gnathodus* of strongly polyphyletic behaviour had already been ever more narrowed down in the years past, namely it has been essentially restricted to the *semiglaber—bilineatus* and *girtyi* groups. Since the type species of *Gnathodus* PANDER sure inough does not coincide with these both groups which could belong to different genera, the name *Dryphenotus* suggested by C. L. COOPER, 1939 will be here assigned to the *semiglaber—bilineatus* group.

3. THE AGE OF THE SAMPLES

Sample 36

According to A. C. HIGGINS, 1975 the *Idiognathoides sinuatus* starts in the RIa (Kinderscoutian, Lower Namurian B, Lower Bashkirian, Earlier Morrowian) and persists up to the Earlier Westphalian (Upper but not uppermost Morrowian, Upper Bashkirian). According to A. C. HIGGINS, 1975 the *Idiognathoides noduliferus inaequalis* starts in the Lower Chokerian, after H. R. LANE et J. J. STRAKA, 1974, however, only in the uppermost Chokerian. So, it occurs first about on the basis of the Pennsylvanian (lowermost Morrowian, lowermost Bashkirian, uppermost

Namurian *A*). The uppermost range of the species as well as also that of the subspecies *I. noduliferus inaequalis* lies at the upper boundary of the Kinderscoutian. Thus, the sample 36 can be assigned to the Kinderscoutian (Lower Namurian *B*, Lower Bashkirian, Lower Morrovian). Since the identification of *I. noduliferus inaequalis* is less exact just because of the fragmentary preservation of the single available specimen and otherwise *I. sinuatus* is represented by highly evoluted forms, the sample 36 may be still a little younger, too, but it belongs by all means to the Bashkirian.

Sample 45

The upper range of *Polygnathus inornatus* (as well as of the genus *Polygnathus* in general) is the Middle Visean (after G. BISCHOFF, 1957 *cu IIIα*). Since *Dryphenotus bilineatus* starts in the Upper *cu IIδ* (D. MEISCHNER, 1970), according to other authors (G. BISCHOFF, 1957) only in the *cu IIIα*, the age of the sample 45 can be regarded very exact as Upper *cu IIδ* to *cu IIIα*.

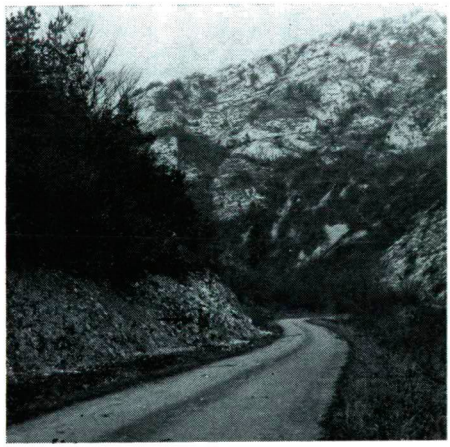


Fig. 2. Dark schists alternatig with grey limestone beds (sample No. 45). In the background the light massive limestones of Uppony.

Sample 40

The abundant occurrence of the index species *Bispathodus costatus* covers unambiguously the *costatus* zone of the Upper *do V* and Lower *do VI.** In the case of some other species further confining of ages can be performed. *Palmatolepis gracilis sigmoidalis* occurs in the upper part of the lower to lower half of the upper *costatus* zone (i.e. uppermost *do V* to lower *do VI*). *Bisppathodus ultimus* starts in the middle *costatus* zone. According to W. ZIEGLER (1969), *Pseudopolygnathus trigonicus* occurs only in the upper half of the middle and in the lower half of the upper *costatus* zone, and after P. PÖLSLER (1969), too, it is restricted in the Carnian Alps to this domain.

Consequently, the conodont fauna of sample 40 can be assigned to the middle to upper *costatus* zone, i.e. to *do VI*. On the basis of the abundant occurrence of

* When following the considerations of H. ALBERTI, H. GROSS-UFFENORDE *et al.*, [1974] and assigning the Stockum Limestone to the Lower Carboniferous, the *do VI* will be ended with the *costatus* zone.

B. ultimus and the absence of *B. jugosus* as well as after the occurrence of *Pseudopolygnathus trigonicus*, both the lower half of the middle *costatus* zone and the upper half of the upper *costatus* zone (i.e. where *Palmatolepis gracilis sigmoidalis* and *Pseudopolygnathus trigonicus* do not occur more) can be excluded (see W. ZIEGLER, 1971).

Fig. 3. Sampling-point No. 40, about 1170 m SE of the barrier at Uppony.

Sample 41

Palmatolepis minuta exists from the uppermost *do I* up to *do IV*, the *P. gracilis gracilis* from *do IIβ* up to *do IV*. From this a classification offers from *do IIβ* up to *do IV*. The identification of the *Polygnathus*-species is uncertain, therefore it is unsuitable for chronological classification. However, it can attest rather an assignment to *do IIβ*.

4. DISCUSSION OF THE NEW CHRONOLOGICAL CLASSIFICATION

The assignment of the limestone—schist complex of the Uppony Mountains to the Upper Devonian to Lower Middle Carboniferous implies a drastic change of the classification when taking into account the recently predominating view on the Silurian age of this sequence (see K. BALOGH–L. KŐRÖSSY, 1974: classification after J. ORAVECZ). On the other hand the former concepts (Z. SCHRÉTER, 1945; K. BALOGH, 1964 and other authors), that the Paleozoic of the Uppony Mountains is the direct floor of that of the Bükk Mountains and is of Tournaisian and Visean age, rather approach the reality. The fact that no evidence of the Tournaisian and Lower Visean age has been found is caused by the very great distances between the samples*. E.g. between the samples 40 *(do VI)* and 36 *(Bashkirian)* no conodont-bearing samples were found. The sample 45 providing the Visean fauna is found between sample 41 of *do II–do IV* age and the underlying massive limestone. The Visean age of the sample 45 evidences that within the limestone–schist complex strong

* An additional sampling near the sample 45 (sample 45A) provided a rich conodont fauna of the Lower Visean *cu IIβ* to *δ* with *Dryphenotus delicatus* BRANSON et MEHL, 1938, *Polygnathus communis* BRANSON et MEHL, 1934 and with numerous fragments of costate *Polygnathus* species.

tectonic movements should be taken into account (the complicated tectonics of the area had been emphasized already by K. BALOGH, 1964), thus its thickness should be estimated smaller as it is given by K. BALOGH, 1964. Overtrustings within the limestone–schist complex can be traced only by means of voluminous microstratigraphic and micropaleontologic investigations. After having results of such investigations the real thickness of the limestone–schist could be determined.

Fig. 4. Sampling-point No. 41, about 570 m SE of the barrier at Uppony, with rose-red and yellow limestones.

Since sample 36 derives from the uppermost limestone bank of the limestone–schist complex immediate below the sandstone–schist complex, its age is a fair starting point to determine that of the latter one. It begins in the (Lower) Bashkirian. Its upper range can be indirectly determined when in the Middle Carboniferous strata of the Bükk Mountains the unambiguously overlying rocks of the sandstone–schist complex of the Uppony Mountains are seen. The oldest fauna of the Paleozoic in the Bükk Mountains can be assigned to the Moscovian. Since below these beds mostly fossil-free schists and sandstones are found which are regarded as the uppermost (though in the Uppony Mountains not exposed) part of the sandstone–schist complex, the sandstone–schist complex will be practically assigned to the Bashkirian (Namurian *B* to Westphalian *B*). In this way the complete coincidence with the age of the Hochwipfel sequence of the Southern Alps can be derived, that proved to be Namurian to Westphalian *B* on the basis of the palynological data of F. FRANCAVILLA, 1966. Consequently, there is a harmony not only between the lithofacies of the Hochwipfel beds and that of the sandstone–schist complex (in both cases dark shales and greywackes, and lydites in the lower part at least), but the age of both complexes is the same, too. The conodont faunas discovered by P. PÖLSLER, 1969 in the limestone of the lower part of the Hochwipfel beds, do not contradict to this; these indicates that the sandy-clayey sedimentation began locally already in the Lower Carboniferous. This is only the question of definition of the Hochwipfel beds and of the sandstone–schist complex, because also the upper part of the limestone–schist complex consisting mostly of dark shales and subordinately of greywackes interlingered by only few thin limestone beds can be correlated with the lower Hochwipfel sequence, in the sense of PÖLSLER and than a

beginning of the shales sandy formation of the Uppony Mountains to the Lower Carboniferous is obtained.

While the upper stratigraphic range of the limestone–schist complex is unambiguous since the sample 36 was taken from the uppermost limestone horizon which is layered already in a prevailingly schistose sequence, the lower stratigraphic boundary of the limestone–schist complex is still not completely cleared. Between the sample 41 and the underlying massive limestone a rather thick sequence of lamellated limestones and schists is found. Even if Lower Carboniferous was intercalated here *(sample 45)*, also Earlier Upper Devonian appears between sample 41 and the massive limestone. In this way, in addition to its Carboniferous part (Lower Carboniferous up to Lowermost Middle Carboniferous) the limestone–schist complex may include also the whole Upper Devonian (undoubtedly proved the Middle and Late Upper Devonian). Thus, the massive limestone belongs probably to the Middle Devonian. Consequently, a classification of the Paleozoic of the Uppony Mountains into the Middle Devonian to Bashkirian can be carried out as against the assignment to the Ordovician and Silurian.

Though, according to our recent knowledge, gaps within the Paleozoic of the Uppony Mountains cannot be certainly excluded, at least no angular discordances are found in the Paleozoic of the Uppony Mountains. Consequently, no Hercynian orogenic movements can be proved between the Middle Devonian and the Bashkirian. Since also in the Bükk Mountains no Asturian and younger Hercynian orogenic movements were detected, the Hercynian orogeny could not have important role in the areas of the Bükk and Uppony Mountains, as well. In this relation the age of metamorphism is very interesting, too. It is commonly accepted that the epizonal metamorphism of the Devonian and Lower Carboniferous strata is of Hercynian age. The metamorphism of the Middle Devonian to Middle Carboniferous strata of the Uppony Mountains and that of the Lower Carboniferous strata of the Gemerides (see H. KOZUR, R. MOCK and H. MOSTLER, 1976) is not stronger than that of the Triassic of the Bükk Mountains, of the Meliata Series and parts of the Triassic in the Rudabánya Mountains, and this fact contradicts to the statement above. Thus, metamorphism should be of Alpine age. Although from Upper Devonian up to the Earlier Middle Carboniferous a typical eugeosyncline sequence with diabases and diabase-tuffs and both in the uppermost limestone–schist and in the sandstone–schist complex flyschlike sediments occur, only germanotype and epirogenic movements without folding and metamorphism passed off during the Hercynian orogeny. In this relation it is interesting that a similar evolution is repeated in the Middle and Upper Triassic, where in the Ladinian and Carnian an eugeosynclinal sequence with numerous basic volcanites (including thick pillow lavas), further deepwater flysch with radiolarites and manganese oxide nodules are deposited. These sequence are overlain without folding and metamorphism by Norian carbonate sequences (the assignment of the "Lower and Middle Ladinian" schist complex to the Carnian and that of the "Upper Ladinian–Carnian" limestone complex of the Bükk Mountains to the Norian is based on the conodont fauna and has been performed by H. KOZUR et R. MOCK, 1977), i.e. by a moderately deep basin facies (cherty limestones reaching up to Sevat) and by thick sequence of shallow water carbonates, similarly to the Southern Alps. The first folding and metamorphism followed only later. The Hercynian unconsolidated parts of the Alpine orogenic belt (see B. ČIRIĆ and H. R. von GAERTNER, 1964) represent the most mobile zones in the Triassic (similar development is found in the internal zone of the Dinarides).

The position of the Paleozoic in the Uppony Mountains within the Alpine orogenic

belt is especially interesting. A. TOLLMANN (1969) considers the peri-Adriatic lineament to be continued south of the Bükk Mountains. Thus, he makes parallel not only the Paleozoic of the Uppony and Szendrő Mountains, but also the Paleozoic of the Bükk Mountains with that of the Northern Alps, with that, however, practically no stronger relations are found (marine Upper Permian in the Bükk Mountains!) H. KOZUR and R. MOCK (1973) compared first the Paleozoic of the Uppony and Szendrő Mountains with that of the South Alpine–Dinaride sequences and they took a near-suture position of the Gemeride–Paleozoic similarly to the Carboniferous of Nötsch–Gailtal. Similarly to H. KOZUR and R. MOCK (1973), H. W. FLÜGEL (1975) considers the Paleozoic of the Bükk Mountains to be of South-Alpine type but he takes the Paleozoic of the Uppony–Szendrő Mountains to be north of the continuation of the peri-Adriatic lineament. The possibility, however, is maintained that the continuation of the peri-Adriatic lineament may draw between the Uppony Mountains and Gemerides (Darnó-line). The recent stratigraphic classification of the Uppony–Paleozoic affirms the suggestions of H. KOZUR and R. MOCK (1973). The Paleozoic of the Uppony Mountains with its typical "Hochwipfel-flysch" can be compared only with the South-Alpine–Dinaride facies and in this relation more stress should lay on the comparison with the Dinarides. The highest similarity is found to the Inner–Dinaride zone west of the Serbo–Macedonian massif and especially to the sequence in the Medvednica Mountains. Here the presumably Middle Devonian massive epimetamorphic crystalline limestone is overlain by likewise epimetamorphic clastic–carbonate sequence of Upper Devonian and Lower Carboniferous which becomes in the later Lower Carboniferous more and more clastic. The Middle Carboniferous is represented by a similarly epimetamorphic clastic sequence and the Triassic is also epimetamorphic, while the Ladinian–Carnian part is very similar to the Paleozoic of the same area (see Z. DURDANOVIĆ, 1973). A perfect similarity seems to be realized with the sequences of the Uppony–Bükk Mountains, based not only on lithological but on the tectonic development, too (absent or insignificant Hercynian orogeny, Alpine epizonal metamorphism). It is also worthy of mention that the areas being highly mobile in the Triassic lie always west of the massifs consolidated strongly during the Baikalian and overprinted by the Hercynian orogeny (Inner–Dinarides west of the Serbo–Macedonian Massif, the Medvednica Mountains west of the "Tisia Massif" in sense of H. W. FLÜGEL, (1975), and the Bükk–Meliata domain west of the Zemplin horst). Thus, the Bükk–(Uppony)–Meliata domain should not be considered by all means to be the branch of the Dinarides, but it rather represents the Inner–Carpathian domain analogue of the Inner–Dinaride domain (in the paper of H. KOZUR and R. MOCK, 1973 it is called South–Slovako–Carpathian, similar concepts are found at M. MAHEL, 1975). A possible connection of the Uppony–Bükk domain, especially in the Paleozoic is the Igal Trench (see H. W. FLÜGEL, 1975) in which the Lower Carboniferous is represented by a facies similar to that of the Gemerides (but by a different one as that of the Upponyian Lower Carboniferous) and also marine Upper Permian (similarly to the Bükk Mountains) occurs. The connection through the Igal Trench (which is to be considered of course in its whole extension as Inner–Carpathian could explain the similarities both to the South–Alpine and to the Dinaride domain and especially to the facies of the Medvednica Mountains. Already in the Upper Permian, however, a single connection through the Igal Trench comes up against some difficulties except one considers it as an unit extremely narrowed during the Alpine orogeny. A connection through a narrow trench (continental Upper Permian both in the Bakony and in the Mecsek Mountains) with shallow water environments

would produce undeniably evaporites in the northwestern part of the trench under the climatic conditions prevailing in the Upper Permian (consequently in the Bükk Mountains, too). On the contrary, the Upper Permian fauna of the Bükk Mountains contains more "Indoarmenian" elements than that of the South-Alps (see K. BALOGH, 1964) where the facies obviously differs a little from the euhaline one.

ACKNOWLEDGEMENTS

For the kind assistance and numerous valuable advices of PROF. DR. K. BALOGH the thanks of authors are expressed.

EXPLANATION OF THE PLATES I—IV

Scale of enlargement for all the specimens: 60x

PLATE I

1—8: *Bispathodus bispathodus* ZIEGLER, SANDBERG et AUSTIN, 1974. — Sample 40 (*1:* 2669; *2:* 2668; *3:* 2671; *4:* 2666; *5:* 2674; *6:* 2670; *7:* 2672; *8:* 2667).
9: *Polygnathus* cf. *inornatus* BRANSON, 1934. — Sample 40 (2693).
10: *Polygnathus* sp. — Sample 40 (2679).
11: *Polygnathus* n. sp. *A.* GROESSENS, 1975. — Sample 40 (2694).
12—15: *Polygnathus communis* BRANSON, 1934. — Sample 40 (*12:* 2652; *13:* 2651; *14:* 2654; *15:* 2653).
16: *Pseudopolygnathus trigonicus* ZIEGLER, 1962. — Sample 40 (2644).
17: *Pseudopolygnathus* n. sp. — Sample 40 (2638).
18: *Bispathodus* cf. *spinulicostatus* (BRANSON, 1934). — Sample 40 (2909).

PLATE II

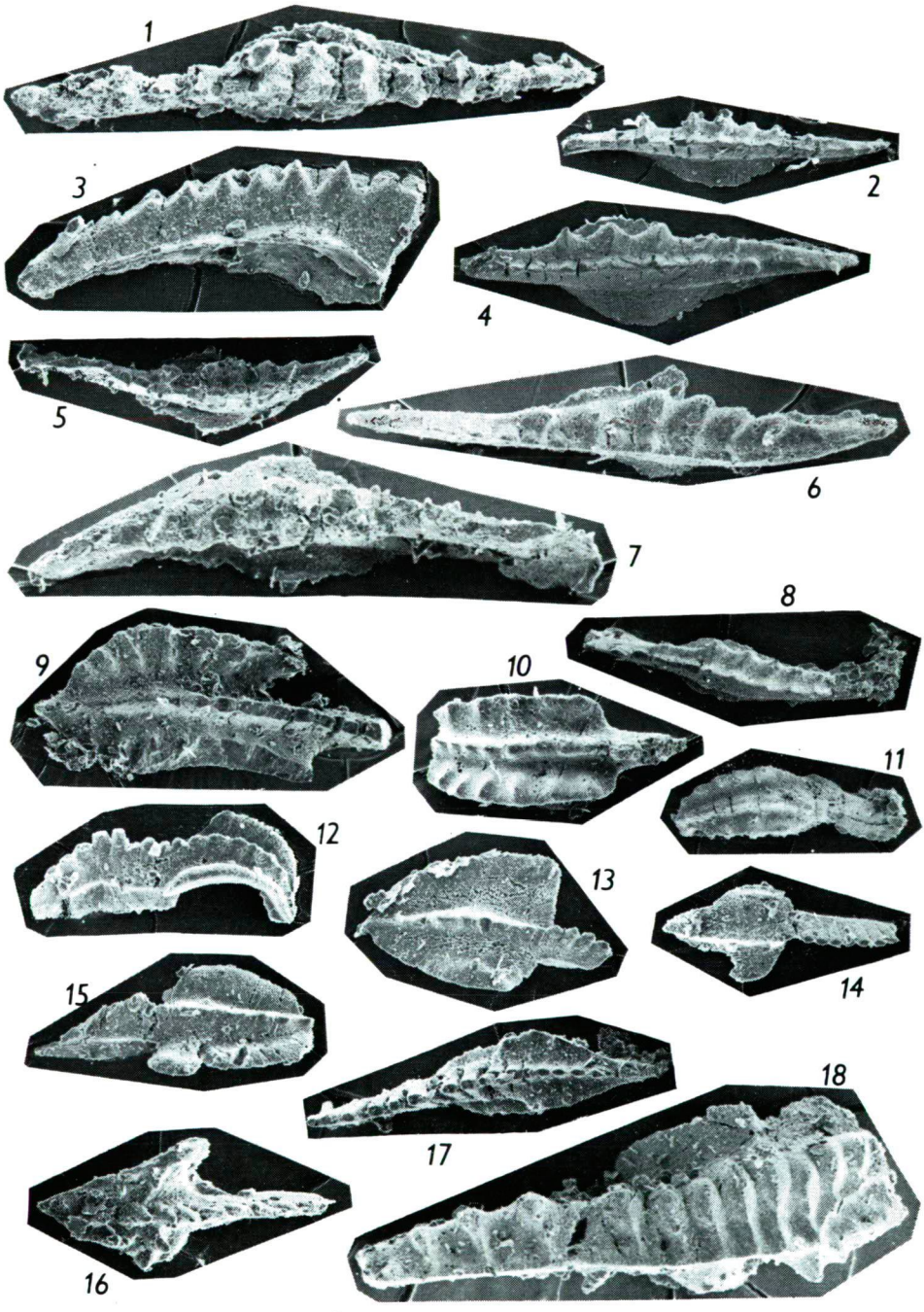
1—8: *Bispathodus costatus* (BRANSON et MEHL, 1934). — Sample 40 (*1:* 2905; *2:* 2899; *3:* 2901; *4:* 2903; *5:* 2902; *6:* 2900; *7:* 2898; *8:* 2904).
9: *Bispathodus costatus* (BRANSON et MEHL, 1934). — Transition to *B. ultimus* (BISCHOFF, 1957). — Sample 40 (2907).
10: *Bispathodus aculeatus* (BRANSON et MEHL, 1934). — Sample 40 (2908).
11: *Bispathodus ultimus* (BISCHOFF, 1957). Transition to *Pseudopolygnathus*. — Sample 40 (2650).
12—17: *Bispathodus ultimus* (BISCHOFF, 1957). — Sample 40 (*12:* 2686; *13:* 2683; *14:* 2681; *15:* 2682; *16:* 2689; *17:* 2690).

PLATE III

1—6: *Bispathodus ultimus* (BISCHOFF, 1957). — Sample 40 (*1:* 2691; *2:* 2687; *3:* 2685; *4:* 2692; *5:* 2684; *6:* 2688).
7—8: *Bispathodus ultimus* (BISCHOFF, 1957). Transition to *Pseudopolygnathus*. — Sample 40 (*7:* 2648; *8:* 2647).
9—11: *Bispathodus* sp. — Sample 40 (*9:* 2896; *10:* 2892; *11:* 2895).
12—17: *Palmatolepis gracilis gracilis* BRANSON et MEHL, 1934. — Sample 40 (*12:* 2641; *13:* 2642; *14:* 2639; *15:* 2649; *16:* 2643; *17:* 2640).
18—19: *Palmatolepis gracilis sigmoidalis* ZIEGLER, 1962. — Sample 40 (*18:* 2677; *19:* 2678).

PLATE IV

1—2: *Palmatolepis gracilis sigmoidalis* ZIEGLER, 1962. — Sample 40 (*1:* 2675; *2:* 2676).
3—7: *Dryphenotus* sp. — Sample 45 (*3:* 2660; *4:* 2662; *5:* 2663; *6:* 2664; *7:* 2661).
8: *"Gnathodus" bilineatus* (ROUNDY, 1926). — Sample 45 (2659).
9: *Dryphenotus delicatus* (BRANSON et MEHL, 1934). — Sample 45A (2646).
10: *Dryphenotus semiglaber* (BISCHOFF, 1957). — Sample 45 (2665).
11: *Palmatolepis gracilis gracilis* BRANSON et MEHL, 1934. — Sample 41 (2897).
12: *Palmatolepis minuta* BRANSON et MEHL, 1934. — Sample 41 (2657).
13—14: *Polygnathus* cf. *lauriformis* DREESEN et DUSAR, 1974. — Sample 41 (*13:* 2696; *14:* 2695).
15: *Nothognathella* sp. — Sample 41 (2658).
16: *? Idiognathoides noduliferus inaequalis* HIGGINS, 1975. — Sample 36 (2906).
17: *Idiognathoides sinuatus* HARRIS et HOLLINGSWORTH, 1933. — Sample 36 (2637).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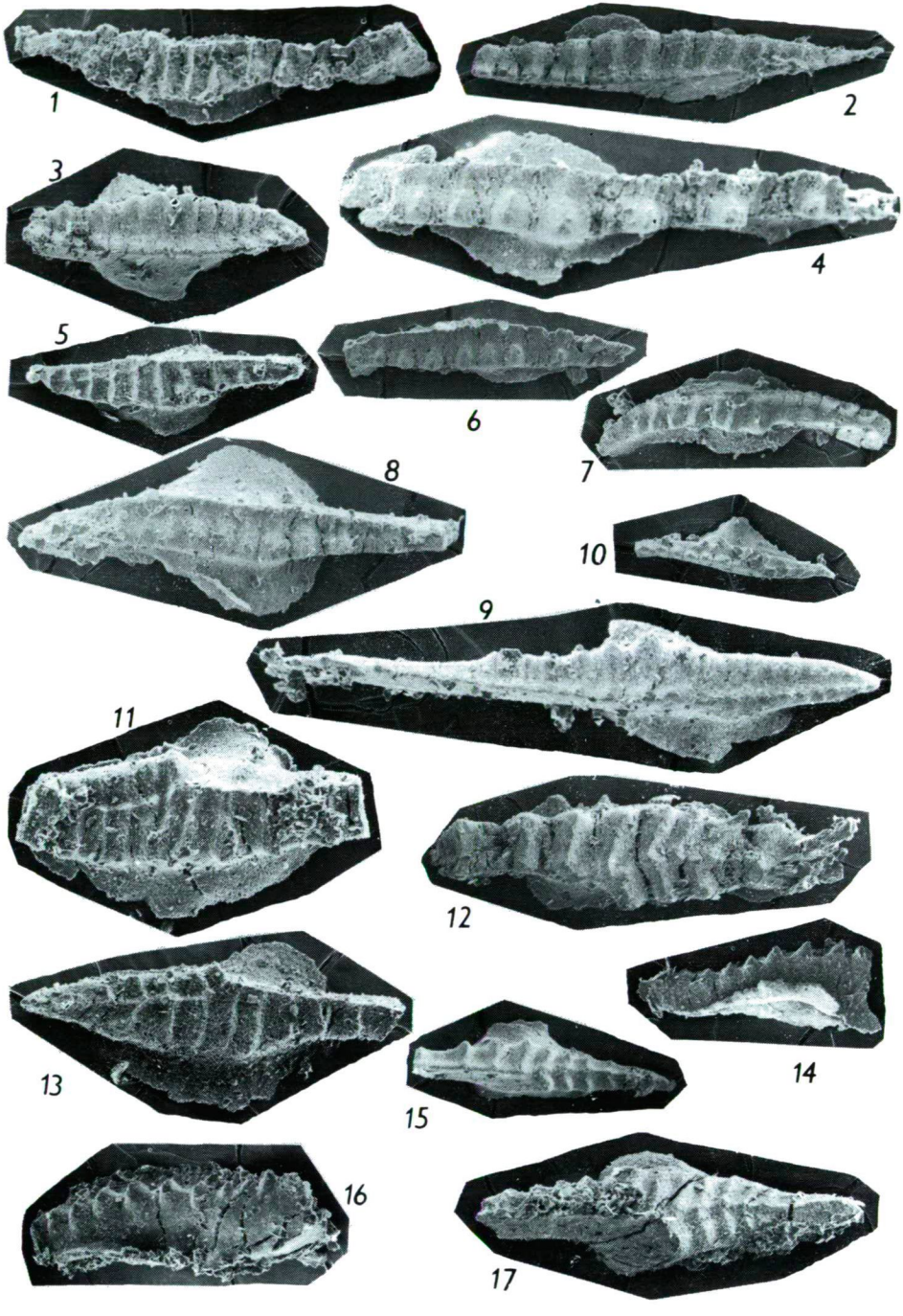
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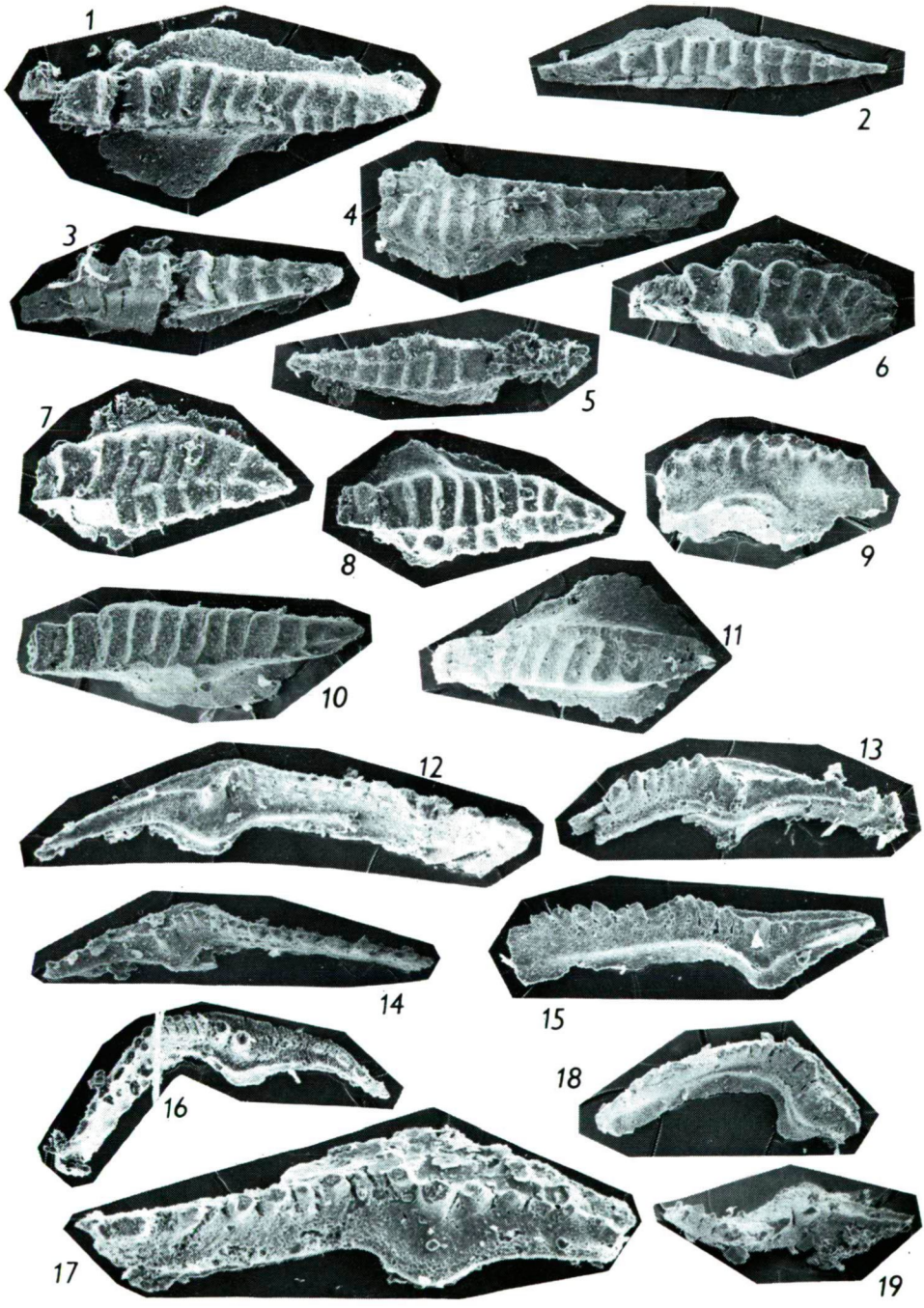
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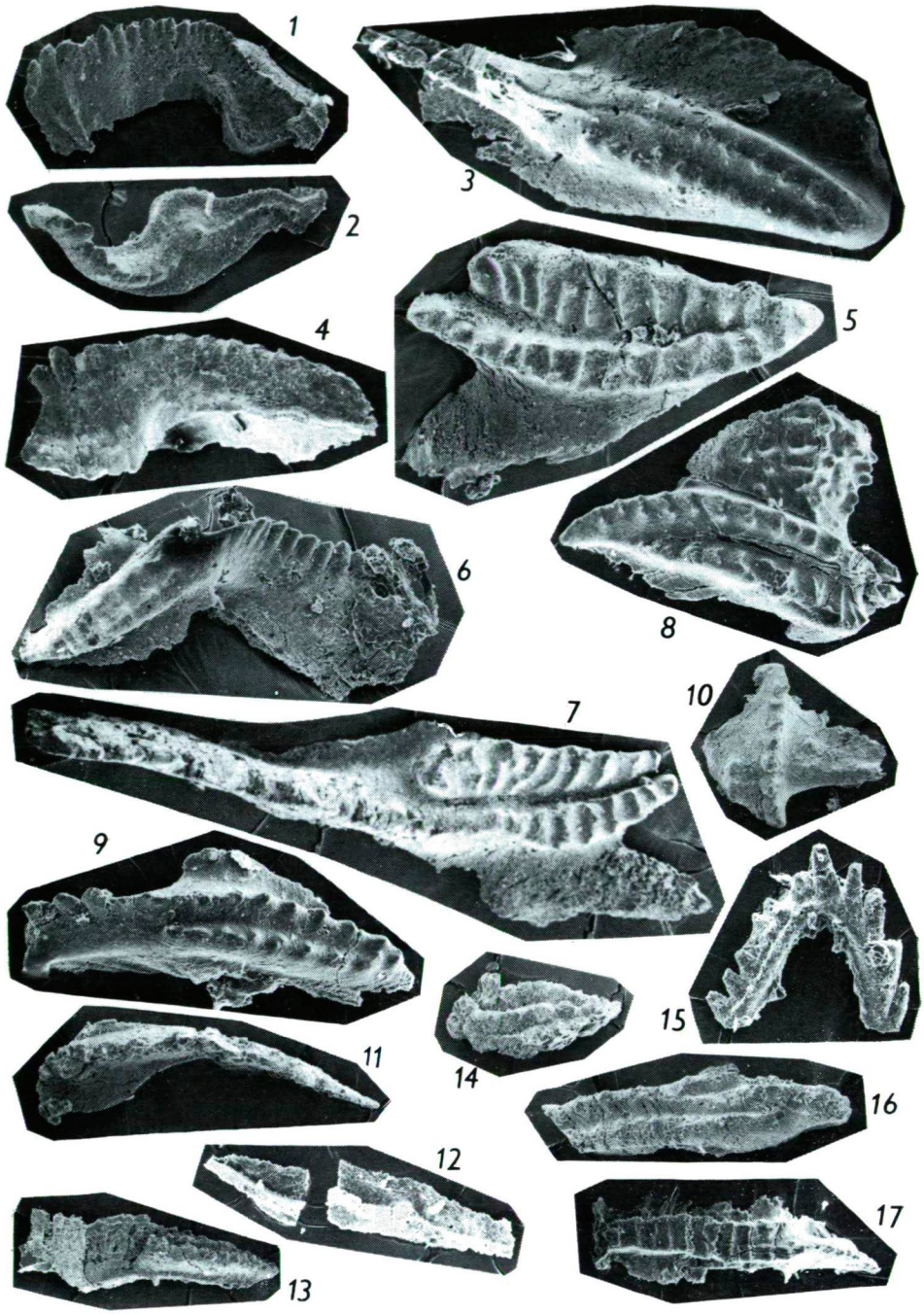
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CONODONTS AND HOLOTHURIAN SCLERITES FROM THE UPPER PERMIAN AND TRIASSIC OF THE BÜKK MOUNTAINS (NORTH HUNGARY)

H. KOZUR and R.MOCK

SUMMARY

5 Upper Permian samples from the Bükk mountains have yielded holothurian sclerites. *Theelia dzhulfaensis* MOSTLER et RAHIMI-YAZD, in the Permian hitherto known only from the Iranian and Soviet Transcaucasia, was found for the first time in the European Upper Permian. 2 Upper Permian samples of the same region have yielded a rather rich conodont fauna with *Anchinathodus minutus* (ELLISON) and *Stepanovites dobruskinae* KOZUR et PJATAKOVA. The age of this fauna was determined to be Upper Baisalian (Dzhulfian).

8 samples have yielded Triassic conodonts. By the aid of these conodonts the age of the cherty limestones of Kiseged could be determined as Middle Sevatian. A Lower to Middle Norian age could be determined for the gray limestones with nodular cherts that overlay the "sericitic shale facies". These cherty limestones were hitherto assigned to the Upper Ladinian (Cordevolian) and Carnian. Therefore the "sericitic shale complex" has not ended in the Middle Ladinian, as hitherto assumed, but it has continued up to the Upper Carnian. A sample immediately above the last shales of the "sericitic shale complex" have yielded an Upper Tuvalian to lowermost Norian conodont fauna. A Julian (Middle Carnian) age could be determined for parts of the "sericitic shale complex" by conodont association of *Gondolella polygnathiformis* BUDUROV et STEFANOV, *Gladigondolella tethydis* (HUCKRIEDE) and *Metapolygnathus* cf. *mišiki* KOZUR et MOCK. The "sericitic shale complex" of the Bükk mountains has therefore the same age as the shaly complex with radiolarites and diabases of the Meliata group. A short summary about the development of the Middle and Upper Triassic in the Bükk mountains and the Meliata group is presented.

The Triassic beds of the Bükk mountains as well as of the Meliata group were deposited in a single highly mobile augeosynclinal belt. After the flysch deposition of Carnian age the deep sea basin was uplifted without any folding and metamorphosis. The Triassic of the Bükk mountains and the Meliata group is very similar to the Dinaric and South Alpine ones and it belongs to the Dinaric faunal province. The austroalpine (North Alpine) Triassic of the Silica nappe must be overthrusted from the north to south over the Dinaric (South Alpine) Triassic of the Meliata group. This is indicated by lithological, tectonical and paleontological evidences.

INTRODUCTION

Resembling to certain parts of the Dinarides, mainly the Ladinian–Carnian complex of the Bükk Mountains (North Hungary) and of the Meliata series (Slovakian Karst) shows extraordinary lithological similarity to the Devonian–Carbo-

niferous sequence. This lithological similarity will be more conspicuous when taking also into consideration the diabases and diabase tuffites occurred both in the Upper Devonian–Lower Carboniferous and the Ladinian–Carnian sequence, as well as their equal metamorphic grade. When taking into consideration the great tectonic complexity of these areas and the usually total absence of macrofossils, it less astonishing that a lot of sequences have been stratigraphically drastically re-classified and numerous classifications are questionable still recently, as well. E.g. the re-classification of sequences assumed formerly to be Carboniferous to the Ladinian by K. BALOGH [1964] in the Bükk Mountains; the re-classification of the Meliata Series from the Permian into the Middle and Upper Triassic by H. KOZUR–R. MOCK [1973a, 1973b]; the frequent re-classifications of the limestone–schist group of the Uppony Mountains, e.g. into the Lower Carboniferous by G. PANTÓ [1954], Á. JÁMBOR [1961], K. BALOGH [1964], into the Silurian by J. ORAVECZ [1965], K. BALOGH–L. KŐRÖSSY [1974], into the Devonian up to Lower Upper Carboniferous by Z. SCHRÉTER [1945], into the Devonian up to Bashkirian by H. KOZUR–R. MOCK [1977]; as well as the re-classification of the „lower sequence" of the magnesite deposits of Ochtiná (Gemerides) into the Upper Visean and Serpukhovian by H. KOZUR – R. MOCK – H. MOSTLER [1976]. The exact stratigraphic classification of the strata is, however, indispensable in order to throw light upon both the tectonics of the area in question and the tectonic of the whole region, as well. Since most strata are free of macrofossils or only a small number of macrofossils of little stratigraphic value are found (e.g. crinoid stem ossicles), first of all the micropaleontological investigations should be taken into account. Nevertheless, the microfossils are also unexpected very sparse in most of the investigated strata. Only the conodonts have a wider spread but they are also less frequent. First the upper stratigraphic reach of the Triassic shale–radiolarite complex was to be determined, of course, in order to confirm its correlation with the corresponding members of the Meliata series, which is, however, also very poor in conodonts. The scarcity of microfossils in this members is caused certainly by the great water depth during the sedimentation. This complex bearing partly manganese oxide nodules and iron jaspilites well as many diabase tuff, furthermore pillow lava intercalations, too, was apparently deposited for the most part below the Triassic lime solving limit. As it was stated by H. KOZUR [1974b, 1976] most of the conodonts have a minimum in deep water and are represented here mostly by juvenile forms. The same phenomenon can be observed in the conodont faunas of this limestone banks within the shale–radiolarite sequence. Difficulties with obtaining of conodonts from epimetamorphic strata originate first of all from the large quantity of the solution residue. On the contrary, disregarding the rather frequent occurrence of an extremely plastical deformation the preservation of the fauna is surprisingly good, thus in most of the samples the conodonts can be easily identified. In this paper only the conodonts and holothurian sclerits from the Upper Permian and Triassic of the Bükk Mountains will be dealt with, but the results of conodont investigations carried out in the Meliata Series will also be used in order to make comparisons with the formers.

LIST OF SAMPLES *(Fig. 1)*

Samples 4, 7, 7A and 10: exposures in the cut of the main road between Eger and Szarvaskő, about 7 km NNW of Eger, in the map of K. BALOGH [1964] called as Ladinian ("dark-grey shale complex with sandstone and cherty limestone intercalations").

Sample 4: collected at the telephone pole No. 64/52 from a grey limestone bed with some chert (2 kg). The banked, partly dolomitic limestone alternates with marls.

Sample 7: at half-way between the telephone poles No. 73/52 and 74/52, marly, strongly folded limestone being very similar to the "Campilian marls" (3 kg). It is underlain by grey limestone.

Sample 7A: Thick bank of grey limestone, one metre above sample 7 (3 kg).

Sample 10: dark-grey to black grained limestone with black nodules of chert (8 kg), occuring within the grey shales between the telephone poles No. 79/52 and 80/52 (15 m of the latter one).

Sample 18: cliff-forming exposure beside the asphalt road between Szilvásvárad—Nagymező and Csipkéskút, about 9,5 km of Szilvásvárad, 550 m behind the kilometre stone 9 KM. The sample derives from the transition of the "Ladinian" shale into "Plateau limestone", from grey banked limestone (4 kg).

Sample 22: Lillafüred, limestone exposure at the cross-way of the main road to Miskolc-Csanyik-völgy. Light grey, banked cherty limestone (being like "Reiflinger-Limestone") which is assigned to the Upper Ladinian—Carnian in the map of K. BALOGH [1964], (3 kg).

Samples 23, 25 and 26: Upper Permian limestone from the Mihalovits-quarry at Nagyvisnyó *(Fig. 2).*

Sample 23: 20 cm thick bank of black limestone with a few crinoid stem ossicles, relatively rich in algae (2 kg).

Sample 25: 40 cm thick bank of black limestone with brachipods, crinoids, algae and nautiloids sp. (2 kg).

Sample 26: 2 metres above the sample 25, black banked limestone with algae, brachiopods, corals and crinoid stem ossicles.

Samples 28, 28A and 30: Upper Permian limestones from the railway cut No. 5 by K. BALOGH [1964] near the hectometre stone 425.

Sample 28: it derives from the upper part of the exposure, from a dark-grey limestone bank full with great brachiopods (2 kg).

Sample 28A: Scattered material around the sample 28, deriving probably from the same layer (3 kg).

Sample 30: 10 cm thick dark-grey limestone bank, 160 cm below the sample 28 (2 kg).

Samples 47—48: Norian limestone on the northern side of the Kiseged hill, east of Eger.

Sample 47: organodetrital banked limestone with large chert nodules, of the two small quarries in the western one, from its lower part (3 kg).

Sample 48: it derives from the eastern quarry, from its lower left side, from a banked limestone, which is like "Reiflinger-Kalk" (3 kg).

III. LIST OF FOSSILS AND AGE OF THE MICROFAUNA

The conodonts and holothurian sclerites are seen in Plates I–III. The scanning photos were carried out with JSM–U3 by K. ŠEBOR and M. ŠVEC in the Dionýz Štúr Geological Institute of Bratislava. The paleontological matter is found at R. MOCK, Bratislava.

Holothurian sclerites

Sample 23, 25, 28, 30:

Achistrum cf. *monochordatum* HODSON, HARRIS et LAWSON, 1956
Achistrum cf. *issleri* (CRONEIS, 1932)

The *Achistrum* species being poor in special peculiarities could not be applied for stratigraphic purposes till now.

Sample 26:

Theelia dzhulfaënsis MOSTLER et RAHIMI-YAZD, 1976

Age: The species ranges from the Abadehian up the Anisian, thus it cannot be applied for exact age determinations. The occurrence of this species in the Bükk Mountains, however, is interesting being known in the Upper Permian only in Iran, as yet.

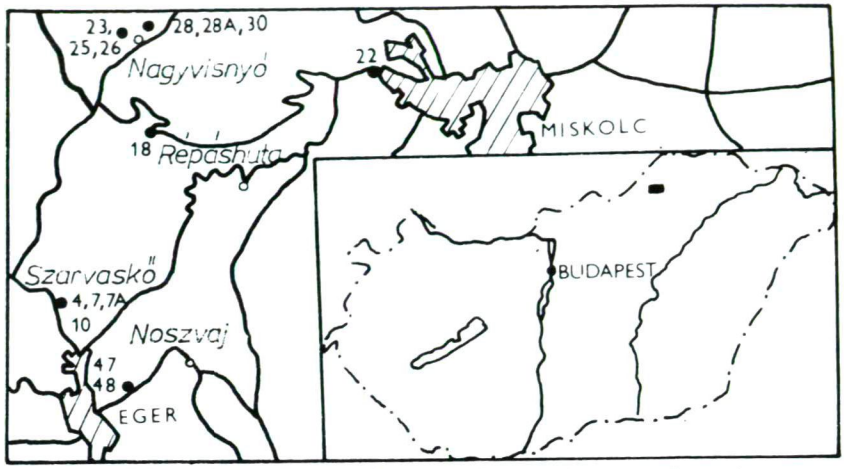


Fig. 1. Schematic sketch on the investigated area and the sampling points.

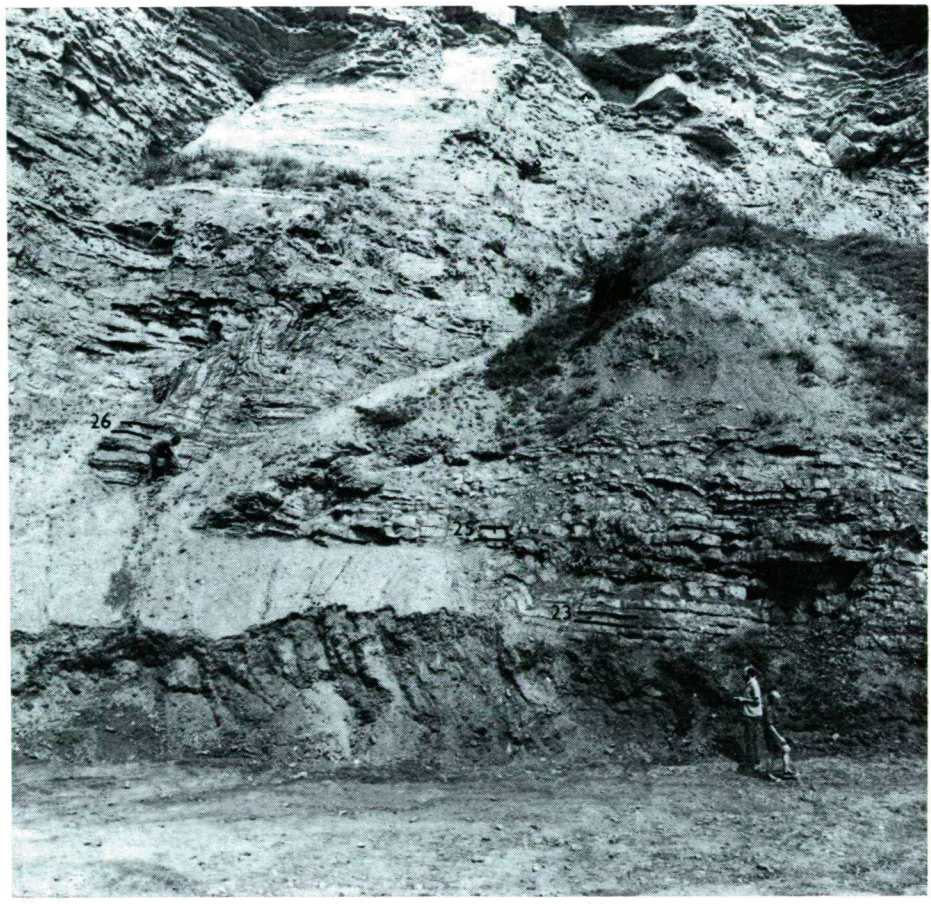


Fig. 2. Mihalovits-quarry in Nagyvisnyó. Lower and middle part of the quarry with the collecting localities of the sample No. 23, 25 and 26. — Photo: R. Mock, 1977

Conodonts

Samples 28, 28A:

Both samples presented the same conodont fauna:

Anchignathodus minutus (ELLISON, 1941), (frequent)
Stepanovites dobruskinae KOZUR et PJATAKOVA, 1975 (rare)

Age: Stepanovites dobruskinae is known only from the Baisalian (Lower Dzhulfian) of Iran and of Transcaucasia. Occurences, which, in consequence of the labile determination, are uncertain taken into the Abadehian, too, and in the Upper Abadehian this species is assumed to occur because in the Lowermost Dzhulfian it is represented already by typical forms. *Anchignathodus minutus* ranges from the Upper Carboniferous up to the uppermost Permian [see H. KOZUR, 1975 and H. KOZUR–H. MOSTLER–A. RAHIMI-YAZD, 1975]. The available forms are highly developed. The rather frequent occurrence of *A. minutus* being extraordinarily rare in the Dorashamian (Upper Dzhulfian) of Iran and Transcaucasia as well as the contemporary absence of *A. julfensis* SWEET, 1973 which is the index fossil of the Dorashamian and the occurrence of *Stepanovites dobruskinae* indicate that the samples 28 and 28A from the fossil-rich horizon of the railway-cut No. 5 of Nagyvisnyó belong still to the Baisalian (because of the highly developed forms of *A. minutus* rather to the Upper Baisalian). In this way a slight difference originates in the classification as compared to that after brachiopods, according to which, on the basis of the occurrence of *Comelicania,* an assignment into the lowermost Dorashamian *(Phisonites–Comelicania fauna)* is rather taken. Since above this brachiopod-rich horizon at the hectometre No. 435 (in the railway cut No. 5 at Nagyvisnyó), after K. BALOGH [1964], there are still about 30–35 m thick Permian sediments there is much to said for the conformity between the Permian and Triassic of the Bükk Mountains suggested by K. BALOGH [1964]. In the case, however, if yet there is a paraconformity (and the profile in the Sebesvíz Valley pictured by S. ANTAL [1975], in which the generally carbonate sequence is interrupted at the Permian/Triassic boundary by a thin slaty marl speaks for this, the gap should be only in the Dorashamian or in the lowermost Triassic. Because of the very high stratigraphic position of the samples 28 and 28A in the Permian proved also on the basis of conodonts, it is excluded that above this horizon still another horizon with *Waagenophyllum* follows as it has been suggested by K. BALOGH [1964]. The opinion of S. ANTAL [1975] can rather be accepted, *i.e.* only one horizon with *Waagenphyllum* is found which is clearly older than the beds from which both conodont-bearing samples derive.

The significance of the conodont fauna demonstrated here is manifested first of all in the fact that within the Tethys itself stratigraphically such high Permian conodont faunas can be found only in a few regions of the world (Iran, Transcaucasia). From the *Bellerophon* strata of the South Alps conodonts are only mentioned buth neither figured nor precisely determined [R. ASSERETO, A. BOSELLINI *et al.* 1973; W. BUGGISCH, 1975; W. BUGGISCH *et al.* 1976].

Sample 4:

Gondolella navicula HUCKRIEDE, 1958 juvenile forms (very rare)
Enantiognathus ziegleri [DIEBEL, 1956], (one specimen)
1 fragment of a compound conodont with *Gladigondolella tethydis* [HUCKRIEDE, 1958].

Age: A more exact classification than into the Upper Anisian up to the Julian is impossible on the basis of the conodonts.

Sample 7:

Gondolella navicula HUCKRIEDE, 1958 juvenile form (one fragment)

Age: Middle to Upper Triassic.

Sample 7A:

One unidentifiable fragment of a compound conodont.

Sample 10:

Gondolella navicula HUCKRIEDE, 1958, mostly juvenile forms (very rare)
Gondolella polygnathiformis BUDUROV et STEFANOV, 1965 (rare)
Metapolygnathus cf. *mišiki* KOZUR et MOCK, 1973 (very rare)
Metapolygnathus pseudodiebeli KOZUR, 1972 (very rare)
Metaprioniodus suevicus [TATGE, 1956] (one specimen)
3 fragments of compound conodonts of the apparatus with *Gladigondolella tethydis* [HUCKRIEDE, 1958].

Age: Julian. — The upper extension of *Gladigondolella tethydis* is Julian. *Gondolella polygnathiformis* occurs only in the Carnian (including the *Frankites sutherlandi*-Zone assigned to the Lower Cordevolian [see H. KOZUR, 1974a]. *Metapolygnathus* cf. *mišiki* is a somewhat more primitive form than the holotype, *i.e.* the platform is somewhat broader and beside the two long and high denticles on the platform there is a tiny third denticle on one side of the platform. Therefore, this form compares to *M. mišiki* as *M. posterus* to *M. bidentatus*. If further matter will be available, this form will be rank as a new species. The deep occurrence of *Metapolygnathus pseudodiebeli* is especially interesting. As it has been demonstrated by H. KOZUR–R. MOCK (in press) this gives a further charasteristic evidences for the Dinaric and Asiatic faunal province. *M. pseudodiebeli* was already described by H. BENDER [1970] from the Julian of Greece and it has been assigned then still to *Tardogondolella abneptis*. E. KRISTAN-TOLLMANN–L. KRYSTYN [1975] found this form also in the Julian. Because of its deep stratigraphic position they created a new species, i.e. *Epigonolella carnica*, although their form demonstrated in Plate 3, Fig. 1. belongs to *M. pseudodiebeli*, while most of the other forms, included also the holotype, belong to *M. nodosus* which similarly in this deep stratigraphic level starts. As it has been suggested by H. KOZUR and R. MOCK (in press) the *M. primitius* taken as yet mostly as synonym of *M. nodosus* [MOSHER, 1968] is an independent species [see also L. C. MOSHER, 1973], which in Tuvalian developed from *Gondolella polygnathiformis* through *M. communisti* HAYASHI [1968]. Among the stratigraphically younger *Metapolygnathus* species E. KRISTAN-TOLLMANN and L. KRYSTYN [1975] discuss only the differences against *M. abneptis* and *M. nodosus* (the latter in the sense of E. KRISTAN-TOLLMANN–L. KRYSTYN, 1975 is *M. primitius*). As it has been emphasized by H. KOZUR–R. MOCK (in press) the differences enumerated against *M. abneptis* cannot be accepted. Thus in the Julian such forms occur that have spiniform elongated marginal denticles (such a form is pictured in *Plate I, Fig.* 22). Nevertheless the absent denticulation of the platform posterior end cannot be used to make difference between the two species as it has been taken by E. KRISTAN-TOLLMANN–L. KRYSTYN, since also the holotype of *M. abneptis* has an undenticulated platform posterior end. There is, however, a difference as against the concept of E. KRISTAN-TOLLMANN–L. KRYSTYN [1975], *i.e.* the contour of *"Epigondolella carnica"* considerably differs from that of *M. abneptis*, the posterior platform part of which becomes considerably and discontinuously broader. As a difference from

M. nodosus [HAYASHI, 1968] E. KRISTAN-TOLLMANN and L. KRYSTYN state that the platform at this species reaches always more than the half-length of the specimen. When they make such a difference to assign the specimen belonging to *M. pseudodiebeli* to another species and figur it as *Epigondolella carnica* in their Plate 3, *Fig. 1*, they come to a contradiction since at this specimen the platform reaches up to the rontal frontal end. The holotype of *M. nodosus* has of course the same relative short platform as that of the holotype of *E. carnica* (see also the expoundings of L. C. MOSHER. 1973 and the forms figurated by H. KOZUR, 1972 in Pl. 4, *Figs 2* and *3* as *M. nodosus* that are near to the holotype of this species). The differences in the development of the carina suggested by E. KRISTAN–TOLLMANN–L. KRYSTYN [1975] (sometimes against *M. primitius* with which they their new species compare) are also not existing. The height of melting of the denticles vary much rather with the ontogenic evolution. In this way no differences can be determined between the *M. nodosus* (in its narrow sense corresponding to the holotype) and the *Epigondolella carnica* if being restricted to the holotype of this species. The specimen, however, demonstrated in Plate 3. *Fig. 4* as *E. carnica* is undoubtedly a new species which can be easily distinguished from *M. nodosus* and from other *Metapolyngathus* species (the holotype of "*E. carnica*" included).

As it has been stated by H. KOZUR–R. MOCK (in press) the *M. pseudodiebeli* develops from the *M. diebeli* (KOZUR et MOSTLER, 1971). By this, the denticulation of the posterior third of the platform will be reduced (such forms are repeatedly pictured by E. KRISTAN–TOLLMANN–L. KRYSTYN as *Epigondolella diebeli*) and an oviform pit will be developed (this latter feature occurs already rather early, *see* e.g. the specimen of *M. diebeli* pictured by H. KOZUR, 1972 in Pl. 2, *Fig. 6.*). Thus it can be stated that the *M. pseudodiebeli* and the *M. nodosus* which start in the Austro-Alpine province first in the Upper Tuvalian, occur already in the Julian in the Dinaric and Asiatic fauna provinces. The theme of the fauna migrations of certain conodont species from the Asiatic resp. Dinaric fauna province into the Austro–Alpine fauna province was dealt with in detail by H. KOZUR–R. MOCK (in press). For this reason the occurrence of *M. nodosus* and *M. pseudodiebeli* may not conduce to assign the sample 10 already into the Upper Tuvalian since both the *M. mišiki* and the *Gladigondolella tethydis* have their upper reach in the Julian.

Sample 18:

Metapolygnathus nodosus [HAYASHI, 1968], (frequent)
Metapolygnathus pseudodiebeli KOZUR, 1972 (rare)
Gondolella navicula HUCKRIEDE, 1958 (frequent)

Age: The absence of *Gladigondolella tethydis* in this conodont-rich sample indicates it to be younger than Julian. In the Lower Tuvalian of the Meliata Triassic *Gondolella polygnathiformis* BUDUROV et STEFANOV predominates far away (J. MELLO–R. MOCK, in press *A*) similarly as it is known from the Austro-Alpine Triassic, where in this period the genus *Metapolygnathus* by no means can be traced till now. In the Middle Tuvalian *G. polygnathiformis* is also very frequent and is accompanied by *Metapolygnathus communisti* HAYASHI. The *nodosus* and *M. pseudodiebeli* are frequent again first in the Upper Tuvalian (*Klamathites macrolobatus*-Zone) and in the Lower Norian (*Mojsisovicsites kerri*-Zone) and at the same time the *G. polygnathiformis* will be strongly restricted here, but it is already absent in the Lower Norian. On the basis of the facts above sample 18 is assigned to the Uppermost Tuvalian up to the Lowermost Norian.

Sample 22:

Metapolygnathus abneptis abneptis [HUCKRIEDE, 1958], (frequent)
Metapolygnathus abneptis spatulatus [HAYASHI, 1968], (very frequent)
Metapolygnathus posterus [KOZUR et MOSTLER, 1971], (rare)
Prioniodina muelleri [TATGE, 1956], (rare)

Age: Later Lower up to Middle Norian. The occurrence of *M. posterus* indicates rather Middle Norian.

Sample 47:

Metapolygnathus bidentatus [MOSHER, 1968], (primitive form, frequent)
Metapolygnathus posterus [KOZUR et MOSTLER, 1971], (rare)
Gondolella navicula HUCKRIEDE, 1958, (mostly juvenile forms, frequent)
Prioniodina muelleri [TATGE, 1956], (rare)
Enantiognathus sp. (one specimen)

Age: Middle *bidentatus*-Zone (Middle Sevatian, *Sagenites giebeli*-Zone)*.

Sample 48:

Gondolella navicus hallstattensis (MOSHER, 1968), (mostly juvenile forms, frequent)
Hibbardella sp. (one specimen)

Age: Norian*.

IV GEOLOGICAL EVALUATION

The period of drastic re-classification of the sequences, assigned formerly into the Paleozoic, into the Triassic is terminated by the imposing Bükk-monograph of K. BALOGH [1964] in the Bükk Mountins and his environs. Only the re-classification of the sequences of the Meliata series (Gemerides, South Slovakia), believed formerly to be Permian or Carboniferous, into Pelsonian up to Norian by H. KOZUR–R. MOCK [1973a, 1973b] falls still to this phase of research, but by means of cono-

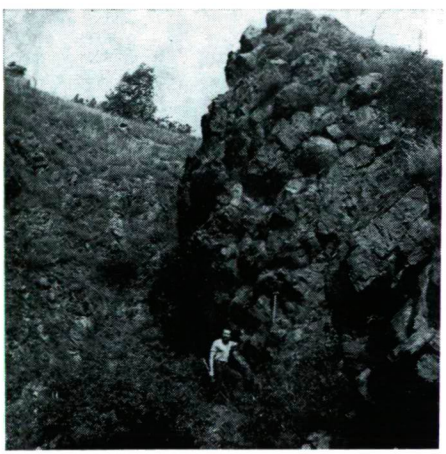


Fig. 3—4: Thick triassic pillow-lavas in the environs of Szarvaskő. — Photo: R. MOCK, 1977

* On the basis of the *Monotia salinaris* BRONN found on the Nagyeged-hill, the rock of these occurrences was assigned already by K. BALOGH, 1964 to the Norian.

donts the biostratigraphically evidenced detailed classification of these strata can be carried out.

To throw light upon the tectonic position of the parts of the Western Carpatians lying south of the Gemeride suture (this suture zone slightly inclining to the south probably transits into the deep-reaching Roznava Line in the depth, *see* P. GRECULA, 1974), the fine-stratigraphic subdivision of the Bükk–Meliata–Rudabánya Trias is as significant as the stratigraphic classification of the pre-Permian Paleozoic of the Gemerides and of the Uppony and Szendrő Mountains. First of all the exact chronological classification of the eugeosynclinal Middle and Upper Triassic of the Bükk Mountains with its spatially and temporally changing of shallow and deep-water-sequences proved to be necessary. Especially the upper stratigraphic limit of the sericitic shale complex, *i.e.* of an epimetamorphic sequence of darkgrey shales and greywackes partly with graded bedding, with intercalations of dark and platy limestones with chert, dark or red siliceous schists containing partly also iron-jaspilites, tuffites and diabases (partly pillow lavas, too, *Figs. 3–4*) had to be elucidated. It was all the more necessary since the previously supposed Lower to Middle Ladinian age of this complex had been occasionally used as the strongest argument as against the identification of the Meliata shales complex with their Bükkian equivalents. The Meliata shale complex has a similar lithofacies as that of the Bükk, but its parts the age of which has been determined until now belong to the Cordevolian and Julian on the basis of conodonts. However, when the correlation of the two complexes is valid it is one of the most decisive arguments to the fact that not only the Triassic of the Bükk Mountains but that of the Meliata Series, *i.e.* the covering unity of the Gemerides, belongs to the South Alpine–Dinaric development. This is evidenced both by the lithofacies and by the basic volcanism of wide extension as well as by the belonging to the Dinaric faunal province [see H. KOZUR, 1973]. On the other hand, this is extraordinarily important in order to make clear the overthrust directions of the Silica-nappe which is to identify with the southern Upper–East–Alpine, and which belongs to the Austro–Alpine (also to North–Alpine) faunal province.

The grey cherty limestones in the direct roof of the sericitic shale complex of the Bükk Mountains were put till now into the Cordevolian and Carnian. The strata of the Répáshuta and Plateau-limestone facies overlying directly the sericitic shales were similarly classified. The sample 22 from the grey cherty limestone facies is, however, unambiguously Norian (Later Lower up to Middle Norian) and the sample 18 which derives from the direct roof of the sericitic shale facies belongs to the Upper Tuvalian up to Lowermost Norian. The limestones from wich the sample 18 was taken and that are connected to the underlying sericitic shale facies through lithological transitions, form at the same time the direct floor of the Plateau-limestones as it has been emphasized already by K. BALOGH [1964]. There is no room for doubt that the grey cherty limestone facies and the Répáshuta limestone facies overlie the sericitic shale facies and on the basis of the macrofauna these strata show similar age than the Plateau-limestone facies.

The "Lower to Middle Ladinian age" of the sericitic shale facies is based on the fact that it directly underlies the above-mentioned three limestone complexes of "Upper Ladinian–Carnian" age. The re-classification of the limestones in the direct roof of the sericitic shale facies into Upper Tuvalian up to Lowermost Norian means, however, expressively that this facies does not come to end in the Middle Ladinian as believed up to the present day but it reaches to the Upper Carnian, as well. The Julian age of a part of the seriticid shale complex can be directly evidenced by the sample

10. In this way the contemporaneity of the Bükkian sericitic shale complex and of the facially similar complex of the Meliata Series is confirmed again. The age of the appearence of the sericitic shale facies and of the corresponding complexes in the Meliata series is hitherto unknown. In the Bükk no samples have been investigated from the zone transitional to the underlying beds. In the Gemerides, at locality Meliata, on the basis of the thick shale and sandstone sequence, an alternation of cherty limestones with shales, siliceous schists and iron-jaspilites occurs which belongs after the recently confirmed classification to the Carnian. In this part of the sequence conodonts occur, the age of which is almost the same as that in the Bükkian sample 10. Thus it seems very probable that the Meliata series forms the direct northern continuation of the Bükk-Triassic. Unfortunately, the shale complex of Meliata lies tectonically on massive white limestones in which intercalations (fissure fillings?) of Pelsonian red limestones with large quantities of conodonts occur. It is unknown whether the limestones and siliceous schists alternating with the dark shales forms already the floor of the shale complex of the Meliata Series or these are only intercalations within this complex. The latter possibility seems to be more probable.

Taking into consideration the results of K. BALOGH [1964] the following schematic profile of the Middle and Upper Triassic of the Bükk Mountains can be demonstrated *(Fig. 5)*. This scheme will be completed by an explanation concerning the age determination of the lower, "Pelsonian" eruptive complex and of an at first proved emergence horizon within the Middle Triassic.

The limestones in the floor of the lower, "Pelsonian" eruptive complex were looked upon formerly as Lower Anisian. Really they are laterally substituted mostly by dolomite, but cannot be considered to be Lower Anisian since *Triadophyllum* occurs in them, and from the Lower Anisian of the Alpine–Dinaric Region no corals are known till now. An assignment of these strata into the Pelsonian seems to be most probable. In the profile of the Sebesvíz Valley the Lower Anisian up to (Lower) Pelsonian dolomite is overlain by a characteristic conglomerate horizon above which first the "Pelsonian" eruptive complex and then the light, probably Ladinian, Fehérkő-limestone follow. Certain parts of the several metres thick conglomerate show typical features of an emergence horizon (the conglomerate is locally interbedded into a lateritic-bauxitic groundmass). Members of this conglomerate horizon in close connection with a dolomite-inclusion-bearing porphyrite agglomerate at the boundary of the dolomite and porphyrite series were first described by K. BALOGH [1964. pp. 379–380, 635] from the western side of the Savós Valley, from the northern side of the Fehérkő cliff and from the Teknős Valley. K. BALOGH assigned an intraformational character to the coarse-grained dolomite breccia. The new, imposing exposures above the Sebesvíz Valley allow to modify the explanation of the phenomenon. Namely, since the eruptive complex consists partly of ignimbrites, it may be of subaerial formation except its uppermost part. Its age may be uppermost Anisian up to lowermost Fassanian and thus it corresponds to the first volcanite maximum of the Southern Alps. The alternation of limestones and tuffites in the transitional zone of the lower eruptive complex towards the Fehérkő-limestone observed already by K. BALOGH [1964] refer to submarine formation conditions concerning the uppermost part of the lower eruptive complex, at least . This part of the lower eruptive complex is certainly younger than the emergence horizon which can be still assigned to the Upper Anisian. The question that the emergence began already in the Lower Illyrian or even in the Upper Pelsonian, or these strata were eroded only in an Upper Illyrian emergence phase, is

recently still open. In connection with the existence of an Upper Anisian emergence horizon it is interesting that the low-grade metamorphic red limestones of Žarnov (at the northeastern end of the Rudabánya Mountains; J. MELLO–R. MOCK, in press *A*) are abundant in their Illyrian part in terrigenic, rolled quartz that seems to derive from the above mentioned emerged land area. The existence of an Anisian emergence phase in the Bükk Mountains shows significant parallelism with the evolution of the Southern Alps.

By means of conodonts from the Lowermost Norian (*Mojsisovicsites kerri*-Zone) up to the Middle Sevatian (*Sagenites giebeli*-Zone) only the age of the gray cherty limestone facies can be evidenced. The Norian age of the Plateau-limestone facies is now still unproved, according to K. BALOGH, however, there are lateral transitions between the chert-bearing grey limestone and the lower part of the Plateau-limestone. The cherty limestone shows partly transitions into the Plateau-limestone through repeated alternation of the beds. The geological evidences suggested by K. BALOGH [1964] for the identity of the floor of the grey cherty limestone facies and of the Plateau-limestone facies seems to be convincing. Thus, within the Norian we have to assume a facies-differentiation to a mostly shallow water facies of Dachstein limestone type and to a basin facies with grey cherty limestones, respectively. Strata younger than Middle Sevatian have not been detected yet. Even if considering only the basin facies being assigned hitherto unambiguously to the Norian, the Norian has, against the former opinions, a wide extension in the Bükk Mountains and in the Trias of Rudabánya [J. MELLO–R. MOCK, in press *A*; S. KOVÁCS, 1977].

The sequence of the Bükk/Meliata Middle and Upper Triassic outlined above shows great similarity to the South Alpine (first of all to the eastern South Alpine) and especially to the Dinaric development. It also faunistically belongs to the Dinaric fauna province [*see* H. KOZUR, 1973; H. KOZUR–R. MOCK, 1973a, b]. The Pelsoniain red limestone (evidenced only in the Triassic of the Meliata and Rudabánya) indicates an early, intensive breaking up of the "carbonate platform". Pelsonian red limestones are known also from the Dinarides. E.g. the red limestones of Han Bulog reach unambigously into the Pelsonian on the basis of the occurrence of *Neospathodus kockeli* (TATGE) [*see* H. KOZUR, 1973]. Areas emerging above the sea level in the Upper Anisian are found also in the Southern Alps. The strong submarine intermediary to mostly basic (first of all in the upper parts) and, rather subordinate, also acid volcanism in the Ladinian and Carnian (starting in the Upper Anisian) is equally characteristic for both the Southern Alps and the Dinarides. So characteristic deep-sea sediments with widespread radiolarites and more scattered iron jaspilites and manganese oxide nodules deposited probably below the Triassic lime-solving limit as found in the Bükk and Meliata Triassic, cannot be seen in the Southern Alps. The similarity is towards the Intra-Dinaric development (Vardar Zone) much stronger.

The Bükk and Meliata Middle and Upper Triassic is an eugeosyncline sequence with strong submarine mostly basic volcanism (diabases, thick pillow lavas, tuffites) and with typical flysch sediments (part of the sericitic shale facies is lithologically very similar to the Kulm-flysch, considering the sediment fabrics, too. After the deposition of the Triassic flysch, however only a shallowing of this area followed without folding and metamorphism (Norian cherty limestones of the basin facies and light shallow water limestones). A similar geosynclinal sequence is found in the Upper Devonian and Carboniferous of this area (Uppony Mountains, see H. KOZUR–R. MOCK, 1977]. The deposition of the eugeosynclinal sequence is connected with

basic volcanites and after the deposition of the flysch, emergence followed without folding and metamorphism.

Finally, let us have some notes on the overthrust direction of the Silica nappe. There is no doubt more regarding both the nappe character of this unit and the direct continuation of the Bükk Triassic northwards to that of the Meliata [*see* also M. MAHEL', 1975]. Within the Bükk/Meliata Triassic there is an interesting succession from south to north. In the Southern Bükk Mountains the proportion of classic sediments is the greatest. Here conglomeratic beds with quartz grains of pea-size, further thick sandstone strata occur that contain terrestrial plant impressions in some places and quite locally very thin coal strips, too [see K. BALOGH, 1964]. On the contrary, in the Northern Bükk Mountains the coarse clastic part is pushed into the background and the limestone intercalations become a little more frequent. In the continuation of this trend towards north, in the Southern Meliata Unit the overlying cherty limestones occur somewhat earlier (Lower Tuvalian; *see* J. MELLO–R. MOCK, in press *B*) and in the northern Meliata Unit the clastic sequence between the under- and overlying carbonates will be strongly reduced and a progressive advance to the development of the Silica nappe in which the clastic discharges are totally absent in the Carnian, occurs. Thus, in the Bükk and Meliata Middle and Upper Triassic the clastic transport came obviously from the south. Consequently, it seems us to be excluded to deduce the Silica nappe from a suture zone lying south of the Bükk Mountains. Similarly, the tracing from a cicatrix south or north of the Uppony Mountains is also excluded. In this latter case the Silica nappe would be originate from the facies range of the Bükk/Meliata Triassic, but this is impossible on the one hand because of the uninterrupted sedimentary environment of the Bükk/Meliata Triassic traceable from the lithofacial development, on the other hand, however, for faunistical reasons (the Silica nappe belonging to the Austro-Alpine fauna province would be surrounded in this case both from the south and from the north by the Dinaric faunal province). In the first case the Silica nappe would be traced from the source area of the clastic Carnian discharges although it holds no clastic sediments in the Carnian. This is all the more incredible since in the Southern Bükk Mountains an apparent south-vergency occurs [*see* K. BALOGH, 1964]. Moreover, a clear contradiction would occur concerning the faunal provinces since in this case the Silica nappe with its typical Austro-Alpine fauna would be settled south of the Bükk/Meliata Triassic characterized by South Alpine–Dinaric fauna. The Silica nappe can be correlated with the Mürztal Facies of the Southern Upper–East–Alpine and it belongs to the same faunal province as the Triassic of the Upper–East–Alpine, as it has been deduced above. Accordingly, there is no possibility either from sedimentological-tectonic or from paleontological points of view to overthurst the Silica nappe onto the Meliata Series from the south. It is far better to suppose a nappe-fan with a southward overthrusting of the Silica nappe, with a near-suture position of the North-Gemeride Triassic as well as with the northwards overthrusting of the Stražov and Choč nappes, which all can be traced from the same suture zone. These conceptions emphasized already by us in 1973 are further firmed by our recent investigations. The only argument against the southward overthrusting of the Silica nappe is that at the Gemeride suture the Gemerides themselves are overthrusted northwards onto the Veporides. It is, however, absolutely not evidenced that this flat overthrust took place simultaneously with the displacement of the Silica nappe, and not later, as it has been suggested here. The southward directed overthrust of the Silica nappe as North-Alpine (better to say North–Slovako–Carpathian, in sense of H. KOZUR–R. MOCK, 1973a, b) unit onto

the South-Alpine-Dinaric (better to say South-Slovako-Carpathian or Intra-West-Carpathian) Bükk/Meliata Triassic contradicts to the common picture in the Alpine-West-Carpathian region. The geological facts, however, cannot be modified in order to obtain conformity with the older conceptions. According to our recent knowledge no feasible choice is expected for southward directed overthrust of the Silica nappe.

ACKNOWLEDGEMENTS

The excursion guided by PROF. DR. K. BALOGH, Szeged, in summer 1977, given us an excellent review on the Triassic of the Bükk Mountains including the most up-to-date knowledges on the researches of its development is gratefully acknowledged here. Further, we are thankful to PROF. DR. E. VÉGH, Budapest, who provided to one of the authors, H. KOZUR, a fair introduction into the geology of the Bükk Mountains during the excursion in autumn 1972.

EXPLANATION OF THE PLATES

PLATE I

Scale of enlargement 60x; excepted Fig. 3 (100x)

1—2: *Achistrum* cf. *issleri* (CRONEIS, 1932). — Sample 23 (2910).
3: *Theelia dzhulfaënsis* MOSTLER et RAHIMI-YAZD, 1976. — Sample 26 (2911)
4—7: *Anchignathodus minutus* (ELLISON, 1941). — Sample 28A (*4:* 2915; *5:* 2912; *6:* 2913; *7:* 2914)
8, 9, 11: *Stepanovites dobruskinae* KOZUR et PJATAKOVA, 1975. — Sample 28 (*8:* 2920; *9:* 2922; *11:* 2921)
10, 12—16: *Anchignathodus minutus* (ELLISON), neoprioniodiniform und enantiognathiform elements. — Sample 28A (*10:* 2917; *12:* 2919; *13:* 2917; *14:* 2916; *15, 16:* 2918)
17: *? Prioniodina (Cypridodella) venusta* (HUCKRIEDE, 1958). — Sample 4 (2927).
18: *Metapolygnathus* cf. *mišiki* KOZUR et MOCK, 1973. — Sample 10 (2925)
19: *? Prioniodina (C.) venusta* (HUCKRIEDE, 1958). — Sample 10 (2924)
21: *Gondolella navicula* HUCKRIEDE, 1958. — Sample 10 (2923).
22: *Metapolygnathus pseudodiebeli* KOZUR, 1972. — Sample 10 (2926)

PLATE II

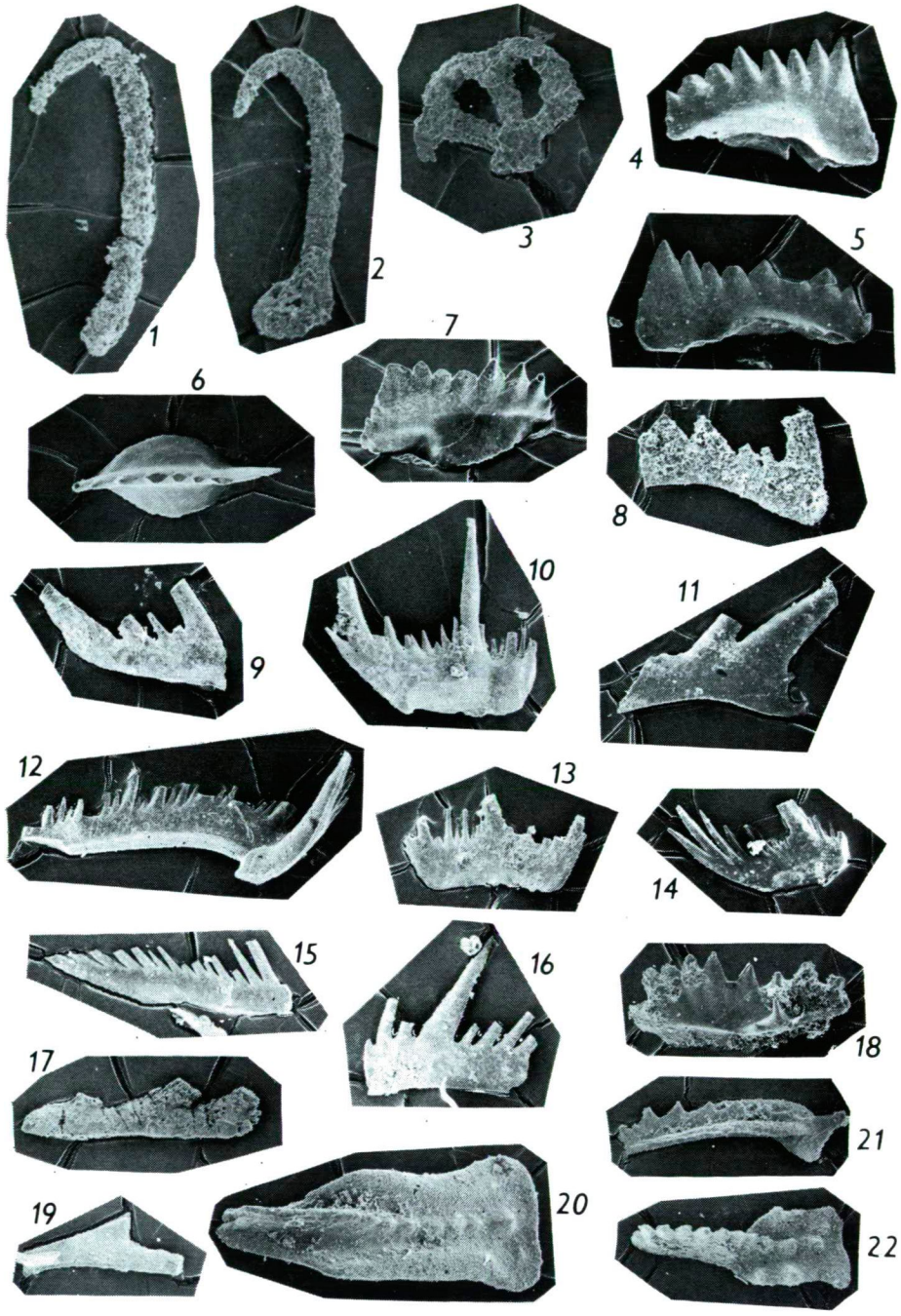
Scale of enlargement: 60x
All the Conodonts from the sample 18

1: *Gondolella navicula* HUCKRIEDE, 1958. — Two entwined specimens (2956)
2—7, 9—11: *Metapolygnathus nodosus* (HAYASHI, 1968). — (*2:* 2952; *3:* 2949; *4:* 2957; *5:* 2951; *6:* 2955; *7:* 2958; *9:* 2954; *10:* 2953; *11:* 2950)
8: *Gondolella navicula* HUCKRIEDE, 1958. — (2959)

PLATE III

Scale of enlargement: 60x

1—12: *Metapolygnathus abneptis* (HUCKRIEDE, 1958). — Sample 22 (*1:* 2946; *2:* 2940; *3:* 2948; *4:* 2939; *5:* 2941; *6:* 2944; *7:* 2938; *8:* 2947; *9:* 2942; *10:* 2945; *11:* 2943; *12:* 2937)
13—16: *Gondolella navicula* HUCKRIEDE, 1958. — Sample 47 (*13:* 2934; *14:* 2929; *15:* 2931; *16:* 2929)
17—18: *Metapolygnathus bidentatus* (MOSHER, 1968). — Sample 47 (*17:* 2935; *18:* 2930).
19—21: *Metapolygnathus posterus* (KOZUR et MOSTLER, 1971). — Sample 47 (*19:* 2930; *20:* 2928; *21:* 2933)
22—23: *Prioniodina (Cypridodella muelleri* (TATGE, 1956). — Sample 47 (2932)
24: *Gondolella navicula hallstattensis* (MOSHER, 1968). — Sample 48 (2936)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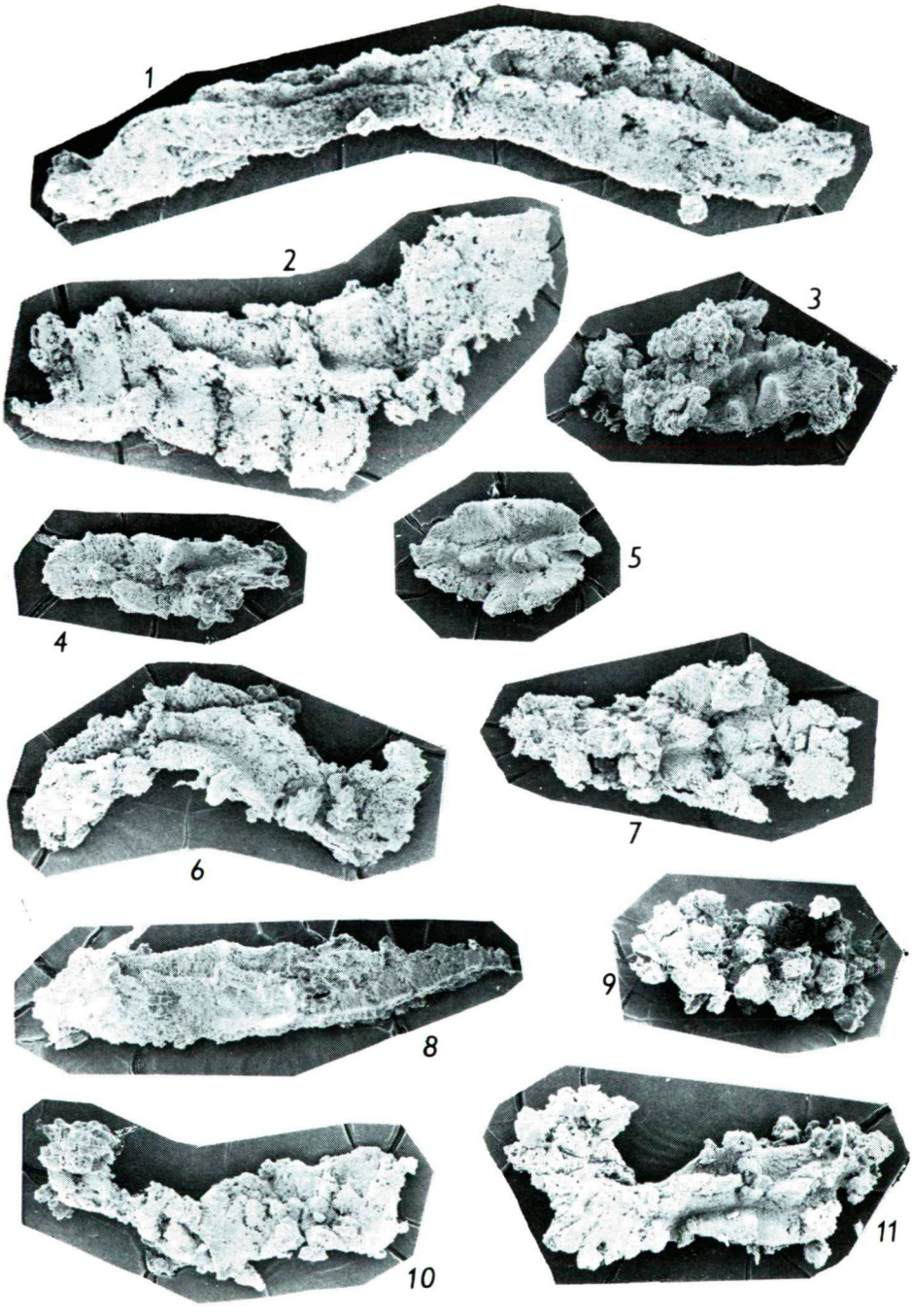
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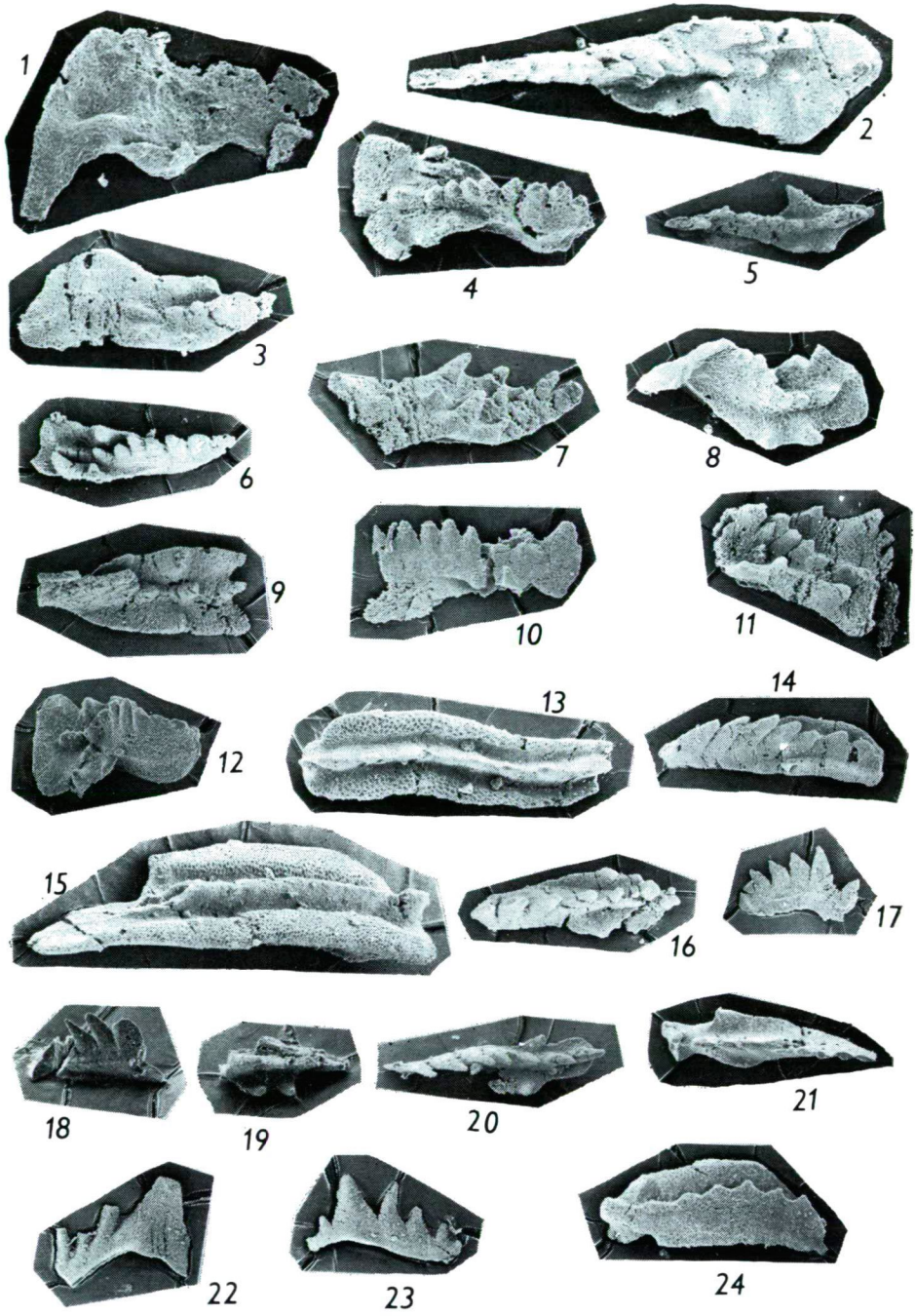
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ORIGIN OF AZAD KASHMIR BAUXITE

KHALIL AHMED MALLICK

ABSTRACT

Bauxite and the associated rocks from seven different localities in Muzaffarabad and Kotli areas of Azad Kashmir have been investigated for their clay and non-clay mineral composition.

Rocks in contact with the bauxite are the fire clay at the base and Eocene shale at the top. The fire clay overlies the precipitation breccia of Permo-Carboniferous age.

No relationship could be established between the precipitation breccia and the bottom of the overlying fire clay. The undulatory contact between the fire clay and the bottom of the bauxite which grades upward into highly pisolitic variety indicates their common parentage in alkaline igneous and metamorphic rocks exposed to the north and north-east of Muzaffarabad Trough. The environment and cycle of deposition of the bauxite and the fire clay were also similar with some break and changes when the pisolites of the bauxites were being formed.

INTRODUCTION

The study of origin of the bauxite whether residual or detrial is not an easy task. The problem becomes more complicated when the contact and the vertical gradation between the bauxite and the underlying rock is not exposed. The difficulty is further enhenced if the bauxite is filling karst depressions. The karst depression filling bauxites were considered as solution residue of limestone till BRIDGE [1950] showed from a study of the bauxite deposits associated with limestone in Georgia and Alabama, ROCH [1956, 1957] from the French deposits, GOLDICH and BERQUIST [1947, 1948] from Haiti and Dominican Republic bauxites and ZANS [1952, 1956, 1959] from Caribbean bauxite deposits in general and Jamaican bauxite deposits in particular, that the source material is not the limestone but some alumino-silicate rocks exposed in the vicinity. Likewise BUSHINSKY [1958] also came to the same conclusion after studying the red bauxites of the Urals. However, it is still a controversial topic and requires more work to solve the problem. The present article comprises a mineralogical study of the bauxite and the associated rocks of Muzaffarabad and Kotli areas in Azad Kashmir which also do not have simple type of contacts. The contact between the bauxite and the underlying fire clay is undulatory and shows vertical gradation into each other. The precipitation breccia underneath the fire clay fills the karst depressions of Muzaffarabad Formation. Hence attempts have been made to determine the source rocks and to reconstruct the depositional environment of the bauxite and the associated rocks in these areas.

It is assumed that kaolinite is stable in low pH range (neutral to acidic) and active leaching with alkalies and alkaline earth removed. The presence of calcium hinders kaolinite formation. Illite is stable in an alkaline environment, and poor leaching with the presence of potassium. Similarly montmorillonite is stable in an alkaline environment and poor leaching with the presence of Mg [GRIM, 1951 and

KELLER, 1956]. Although the complex situation in nature will not make this assumption rigorously true for each bauxite deposit, the over all effects as judged from many samples may give a good indication of the composition of the parent rock.

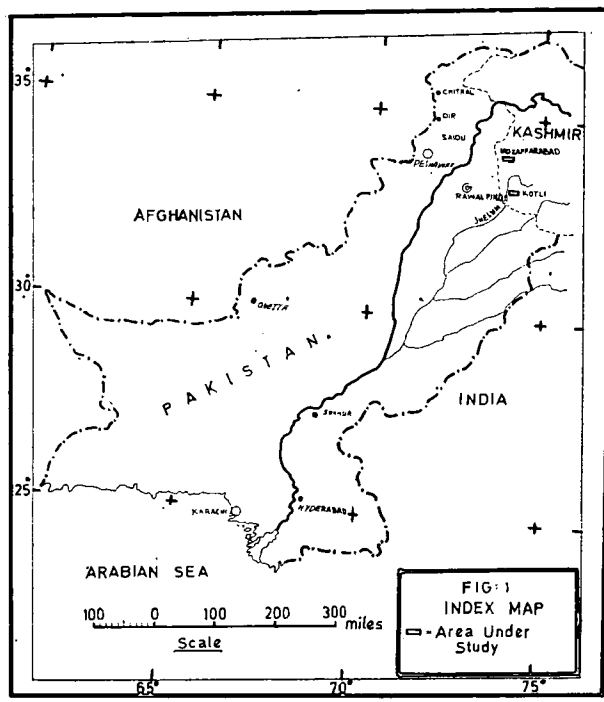
The presence of minerals mentioned above is expected to indicate consistent mineralogy or a range of mineral variation for a particular rock subjected to bauxitization, provided there is no contamination through other sources. Any radical change in the clay minerals would be a cause of variation like the change in chemical environments and rock contamination of different composition.

The clay minerals as determined in the bauxite and the associated rocks by DTA method show that kaolinite is common in fire clay and the bauxite. The Eocene shale overlies the bauxite and contains kaolinite and halloysite. The precipitation breccia underneath the fire clay shows the presence of quartz only. Hence the clay minerals can be good indicators for distinguishing between the bauxite and the associated rocks of the various localities, as well as in determining the degree of correspondence between them.

Likewise similar and dissimilar non-clay minerals of the bauxite and the associated rocks were studied microscopically to determine the mineralogical correspondence between the composition of the bauxite and the associated rocks. This study is also expected to help in tracing the source rocks of the bauxites.

GENERAL GEOLOGY

General geology of these areas has been described in reports from time to time but no petrographic or mineralogical work on the bauxite and the associated rocks with the above objectives in mind has yet been done.



The localities discussed in this paper are Batmong and Khilla in Muzaffarabad and Sawer Guniamalni, Khander, Kamroti, Nikial, and Salhun in Kotli Thesil of Azad Kashmir territory *(Fig. 1)*. The rocks exposed in the areas of study are Muzaffarabad Formation, the fire clay, the bauxite and the Chhalpani Formations in their chronological order.

Muzaffarabad Formation of Permo-Carboniferous age consists of limestone, dolomite and precipitation breccia. The limestone is thinly bedded, light gray to mottled gray and white in colour, fine grained, hard and partly dolomitic. Joints and fractures are common.

Above the limestone is the dolomite and the top is precipitation breccia. The dolomite is thinly bedded, dark gray to black, hard compact, brittle, and wheathers to gray or dark gray. The precipitation breccia is colourless to white, hard and compact and very resistant to weathering.

The fireclay and the bauxite are Palaeocene in age and overlie the precipitation breccia of Muzaffarabad Formation. The bauxite is light to dark gray, yellow, brown and dirty white in colour and occurs in small pockets and lenses in association with the basal bed of gray and dark gray fire clay. The top of the bauxite bed is pisolitic and grades into non-pisolitic at the bottom. Average thickness of the pisolitic bauxite is 0.79 m and that of the underlying fire clay is 1.5–3 m. At some places the pisolitic bauxite has been completely eroded and only the poorly pisolitic part at the bottom is exposed.

The Chhalpani Formation of Eocene age is composed of dark carbonaceous light gray to olive and yellowish colored calcareous shales; light gray medium grained sandstone; and dark gray to almost black nummulitic limestone. The dark carbonaceous shale of Eocene age above the pisolitic bauxite contains the coal seams.

The main structure in Muzaffarabad area is a recumbent anticline in the east of Muzaffarabad proper. The regional structure in Kotli area is a pitching anticline, but not very tightly folded and turned into a recumbent fold as in Muzaffarabad area.

CLAY MINERALS IN THE BAUXITES AND THE ASSOCIATED ROCKS

Table 1 shows the vertical column of the major rock types, their stratigraphic succession and the types of clay minerals as determined by the differential thermal analysis method.

Fire-clay 1 & 2 and bauxite 1 & 2 (Table 1) represent the bottom and top of the respective beds. Megascopically the bottom of the fire clay can be distinguished from the top mainly by its color. The bottom of the bauxite bed is dark gray and poorly pisolitic while the top of the bed is dark gray to black and highly pisolitic.

Separate samples were collected from the top and the bottom of the beds to study the differences and similarities in clay mineral composition within the bed and also between the beds which appear to be conformable in the field.

The precipitation breccia appears to be very consistent in its chemical composition and no other mineral but quartz is present in each locality. In Kamroti, however, precipitation breccia is missing and the bottom of the fire clay is apparently in contact with the dolomite of Muzaffarabad Formation (Table 1).

The thin sections of the Muzaffarabad dolomite show clustering of grains which is usually more pronounced in the coarser grains and exhibits granular texture. The cementing material between the grains is scanty and the grains lie in random orientations. Conspicuous partings and cleavage directions are present

(Fig. 2a). In some of the grains rhombic cleavages are quite pronounced. It appears that the dolomite is product of inter-action between the limestone and the sea water.

In thin sections of precipitation breccia the grains of quartz appear to dominate the rock body. The cementing material between the grains is very scanty and the texture is quartzitic *(Fig. 2b)*. Few grains of orthoclase and albite showing kaolinization and sericitization effects were also found.

TABLE 1

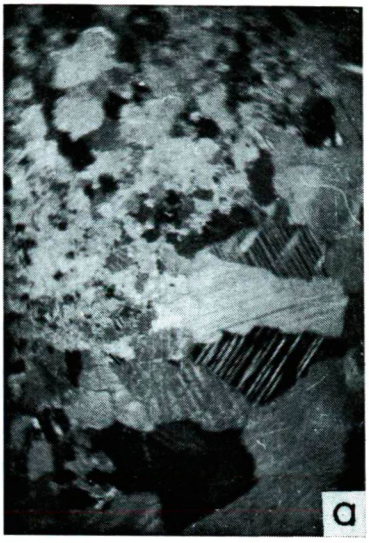
Clay minerals in the bauxite and the associated rocks

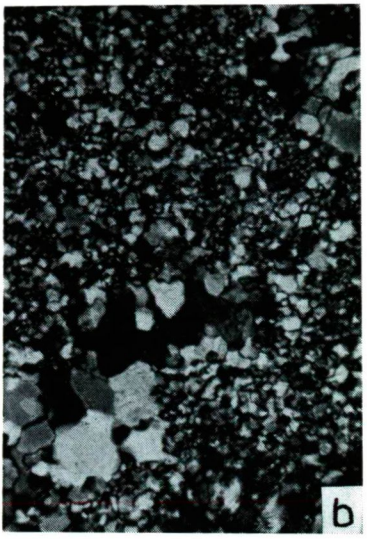
Stratigraphic succession	Localities					
	Batmong	Sawer Gunimalni	Kamroti	Nikial	Khander	Salhum
Eocene Lms.	—	Calcite	Calcite	Calcite	Calcite	
Eocene Sh.	Halloysite	Halloysite	Halloysite	—	Kaolinite	
$Bauxite_2$	Kaolinite	Kaolinite	Diaspore & Kaolinite	Diaspore or Boehmite	Kaolinite	
$Bauxite_1$	Kaolinite	Diaspore or Boehmite	Kaolinite	Kaolinite	Diaspore or Boehmite	Diaspore or Boehmite
Fire $Clay_2$	Kaolinite	Diaspore or Boehmite	Raolinite	Kaolinite	Kaolinite	Kaolinite
Fire $Clay_1$	Kaolinite	Kaolinite	Illite	Diaspore or Boehmite	Diaspore or Boehmite	Diaspore or Boehmite
Muzaffarabad Precipitation Breccia	Quartz	Quartz	—	—	Quartz	Quartz
Muzaffarabad Dolomite	—	—	Dolomite	—	Dolomite	—

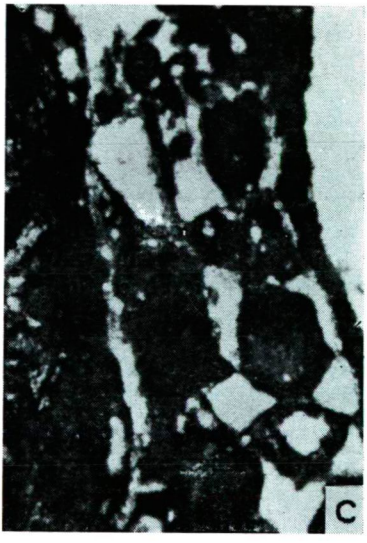
At Khander lateritic patches lie between the precipitation breccia and the fire clay. The laterite is reddish-brown to dark brown in color and exhibits coarse to medium grained texture. No minerals could be identified in the hand specimens due to coatings of iron oxide on the grains.

The thin sections of the samples of laterite show the dominance of orthoclase and albite grains embedded in ferruginous matrix. Some grains of plagioclase and quartz were also found but they are not very common. Usually the grains of feldspars are angular to subangular in their shapes. Most of the grains are kaolinized and sericitized along the margins and cracks but the degree of alteration in feldspar grains present in this section of the laterite samples *(Fig. 2c, d)* show kaolinization effect very conspicuously due to diagenesis.

The study of the thin sections of the fire clay show the presence of the grains of quartz, kaolinized and sericitized albite and orthoclase embedded in blackish brown argillaceous matrix. Few grains of ferromagnesian minerals of pyroxene and amphibole groups were also found in altered condition. The alteration products of these minerals are biotite, epidote and some clay minerals which appear dirty white and cloudy in thin sections *(Fig. 2e)*. Flakes of biotite, few grains of apatite,







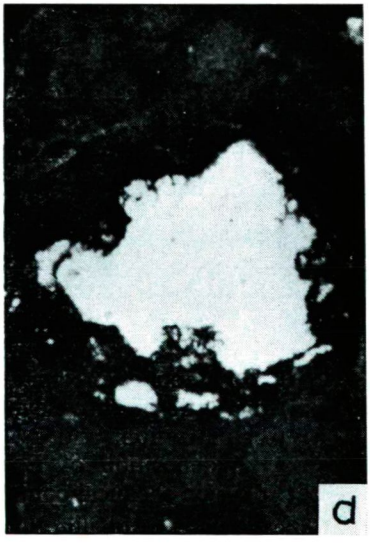


Fig. 2.

a) Fine to coarse grained aggregate of dolomite crystals from Khander, Azad Kashmir 50.4 X, crossed polarizers.
b) An aggregate of anhedral crystals of quartz in precipitation breccia, from Khilla, Azad Kashmir, 50.4 X, crossed polarizers.
c) Feldspar grains embedded in ferruginous matrix of laterite from Khander, Azad Kashmir 50.4 X, polarized light.
d) Kaolinized feldspar grain surrounded by ferruginous matrix of laterite from Khander, Azad Rashmir, 50.4 X, polarized light.

anatase and epidote were also seen under the microscope. Exact identification of the mafic minerals is not possible due to intense decomposition of the mineral grains. However, their outline, the development of craks and the black specks of iron oxide in the body of the grains are quite pronounced. Calcite and muscovite are common. Small specks of iron oxides with random distribution are of common occurrence in the main body of the rock. In all the samples of fire clay from the various localities of Muzaffarabad and Kotli Tehsil, banded structure is quite common and well pronounced *(Fig. 2f)*.

The thin sections of bauxite samples show the presence of argillaceous and ferruginous matrix in which grains of orthoclase, albite, quartz, flakes of muscovite and biotite are embedded. Few grains of anatase, apatite and calcite were also found. Specks of iron oxides were also seen under the microscope.

The grains of albite and orthoclase are angular to subangular in shape. These mineral grains are kaolinized and sericitized though their outlines are quite prominent. Generally in thin sections of the pisolites the mineral grains show their outlines quite conspicuously, but in some cases the grains are so intensely decomposed that the study of the shape is difficult *(Fig. 2g)*. In some of the pisolites few laths of feldspars appear as trapped grains in carbonaceous and argillaceous matrix. In others the rim is lined by altered grains of feldspars, and the internal material is carbonaceous to argillaceous with few grains of decomposed feldspar. In such pisolites the pisolitic structure is not well developed. Some of them exhibit random setting of non-clay mineral grains specially feldspars in them.

In non-pisolitic bauxite samples also some grains of feldspar and other minerals mentioned above, are present, exhibiting the same degree of decomposition of the mineral grains as in the pisolites. In some cases the altered feldspars appear to dominate over the matrix *(Fig. 2h)*.

ENVIRONMENTAL INTERPRETATION

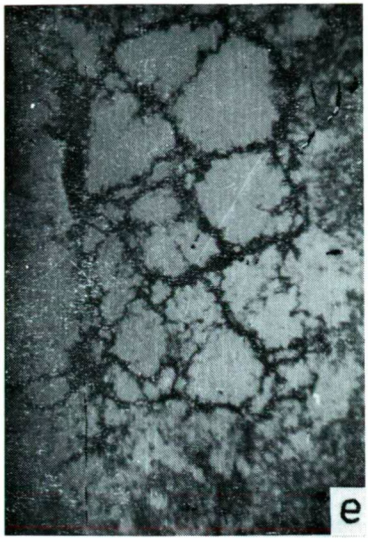
The common occurrence of dolomite in the middle and precipitation breccia at the top of the Muzaffarabad Formation, kaolinite in the fire clay and the bauxite beds, halloysite in Eocene shale and calcite in the limestone alternating with the shale in Azad Kashmir area, suggests that there were at least five phases of sedimentation, namely:

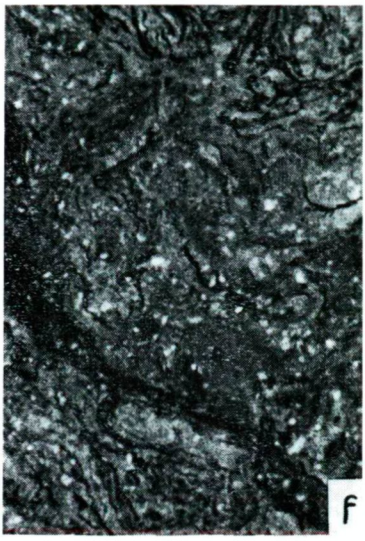
i) dolomitization of Muzaffarabad limestone,
ii) formation of precipitation breccia,
iii) deposition of fire clay,
iv) bauxitization, and
v) the deposition of Eocene shale interbedded with limestone in varying physico-chemical controls of the environment.

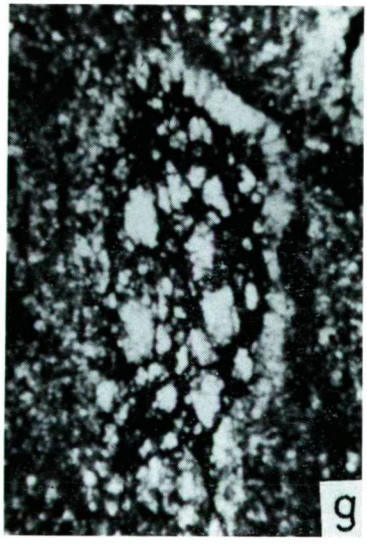
The occurrence of illite in Kamroti locality only can not be explained on the basis of observations of MILNE [1958], BURST [1958] and WHITEHOUSE [1965] who reported the formation of illite from montmorillonite in marine condition, because no evidence of marine deposition of the fire clay of Azad Kashmir was found during the present investigations.

The presence of illite only at the bottom of the fire clay of Kamroti locality can be explained as detrital or derived from muscovite or montmorillonite due to petrodiagenesis.

WEISSE [1948] suggested that gibbsite generally occurs in deposits of Tertiary age, boehmite in deposits of Mesozoic age and diaspore in deposit of Paleozoic age.







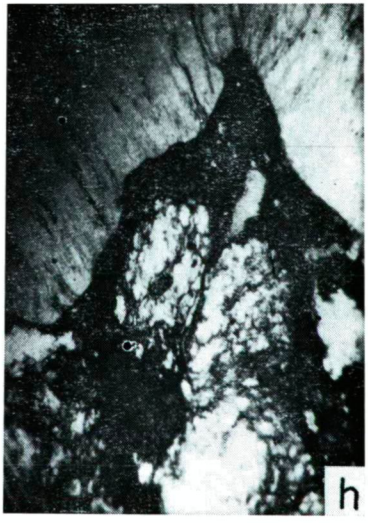


Fig. 2.

e) Decomposed amphibole grains embedded in argillaceous matrix of fire clay from Khander, Azad Kashmir, 50.4 X, crossed polarizer.

f) Banded algal structure with small feldspar grains in the fire clay from Khander, Azad Kashmir. 50.4 X, polarized light.

g) Pisolite of bauxite with decomposed feldspar grains from Batmong, Azad Kashmir, 80.0 X, polarized light.

h) Kaolinized feldspar grains trapped in argillaceous matrix of non-pisolitic bauxite from Khilla, Azad Kashmir, 50.4 X, polarized light.

This age relationship shows that with the geologic time gibbsite is dehydrated and altered to boehmite and boehmite is further altered to diaspore. Increase in pressure and temperature above those that exist at the surface does not appear to be necessary for the formation of these minerals.

BRACEWELL [1962] states that boehmite and diaspore may also be formed by the thermal metamorphism of gibbsitic bauxite or laterite.

J. DE LAPPARENT [1936] considers that boehmitic bauxites are formed at the ground water-table in the presence of humic acid water level and diasporic bauxite below it, where a higher geothermal gradient prevails.

The absence of gibbsite and random distribution of boehmite or diaspore in the bauxite and the associated fire clay of Azad Kashmir make the explanation given by WEISSE [1948] a paradoxical one. It is possible that all the gibbsite present at the beginning of sedimentation has been dehydrated into diaspore or boehmite due to intense tectonic movements to which those rocks were subjected during various phases of Himalayan uplift.

It is likely that the diaspore or boehmite are among the detrital sediments, and so the distribution of these minerals is random. But this seems less probable because diaspore and boehmite are usually the products of petrodiagenesis.

The thermal origin of boehmite and diaspore as suggested by BRACEWELL [1962] does not seem possible in case of the Azad Kashmir bauxite because no field indications or petrographic evidence of igneous activity were found in the bauxite and the associated rocks.

The formation of boehmite and diaspore in Azad Kashmir bauxite and fire clay seems compatible with the observation of DE LAPPARANT [1936] that the action of ground water together with humic components in the water draining from neighbouring peaty forests acted upon the gibbsitic bauxite and produced boehmite and diaspore. The humic component in Azad Kashmir bauxite could have been added from the carbonaceous material which is now present as a coal seam in Eocene shale above the bauxite lenses. Besides, the whole area is covered with pine forest and so the decomposition of the pine trees keep adding humic acid to the bauxite body.

The occurrence of halloysite together with kaolinite in the Eocene shale favours the acidic and oxidizing physico-chemical control of the environment. Therefore, it is further confirmed that acidic environment was responsible for this mineral assemblage.

The formation of clay minerals in the bauxite after the removal of non-aluminous materials is a complex process and it is not well understood. The dominant clay minerals in the fire clay and the bauxites of Azad Kashmir are kaolinite and boehmite or diaspore (Table 1). The kaolinite present in the fire clay and the bauxite is probably a transported material. The result of the present study is compatible with the conclusions reached by PATTERSON [1967], who described extensive transported kaolin deposits underlying or associated with some of the bauxites in Arkansas, the coastal plains of Georgia and Alabama, Guayana and Surinam.

The process of resilicification for the formation of kaolinite as suggested by BUSHINSKY [1964] does not seem to be very active in case of Azad Kashmir bauxite, because halloysite a member of kaolin family is present in Eocene shale which overlie the pisolitic bauxite. If there had been an active process of resilicification then the top of the pisolitic bauxite and the Eocene shale would have shown less percentage of silica and presence of clay minerals like gibbsite, boehmite or diaspore only. But such indications were not found in any of the localities of the present study. Besides, the chemical analyses of the bauxite and the fire clay samples do not show any appreci-

able change in their silica content from the top to the bottom of either beds [MALLICK and VALIULLAH, 1974].

The absence of gibbsite and occurrence of boehmite or diaspore in the fire clay and the bauxite is probably due to the effects of tectonic disturbances caused by Himalayan orogeny which changed gibbsite into the two minerals mentioned above through the process of dehydration and structural compaction.

Besides the evidence based on the study of clay minerals, the presence of altered angular to subangular grains of feldspars, ferromagnesian minerals probably of amphibole group, the quartz grains in the argillaceous matrix of the fire clay and the pisolitic bauxite reveal their transported nature and a common source. The presence of lateritic patches containing comparatively fresher grains of minerals than in the fire clay and the pisolitic bauxite, and its position between them further indicate immature state of the material which formed the two beds.

The negative correspondence of the fire clay and the pisolitic bauxite with the associated underlying precipitation breccia of Muzaffarabad Formation is clearly indicated by the microscopic examination of the thin sections.

The banded structure is probably an indication of the transportation of algal material along with the argillaceous sediments in the basin of deposition. It is also possible that the algal material was present in the basin of deposition, and so, when the transported sediments came in contact with the algal materials, both were deposited together and formed the bed of the fire clay.

It appears from the study of the internal structure of the pisolites that the local disturbances in the environment of bauxitization and pisolite formation were active and caused irregularities in the internal structure and setting of the mineral grains in them. Probably the disturbance in the supply of organic matter and the sediments were more intense at the time of bauxitization.

The provenance of the fire clay and the pisolitic bauxite was most probably in the north and north east of Muzaffarabad Trough which is occupied by the gneisses, schists, slates and the early Tertiary instrusions of granite, syenite, granodiorite and diorite. The positive correspondence between the fire clay and the pisolitic bauxite, on the basis of their mineral assemblage is probably due to their common provenance. The cycle of deposition also appears to be the same with some fluctuations in sedimentation and shallowing of the depositional basin when the pisolitic bauxite was in the process of formation.

While studying the bauxites from occupied Jammu province, ASHOK [1967] concluded that the provenance of the bauxites is in Pir Panjal Trap which is situated further north from the localities of the bauxite deposits. But the present study reveals that the source rocks of Azad Kashmir bauxites are the alkaline igneous and metamorphic rocks as mentioned above and is discussed below:

GEE [1957] is of the opinion that the basin structure of West Pakistan was emerging from the north and was submerging in the south during Mesozoic and Tertiary times. This statement further supports the possibilities of the source rocks for the fire clay and the bauxite deposits to be in the north.

The dominance of feldspar grains in the fire clay and the bauxite samples as compared to silica is probably because of the dominance of alkaline rocks in the provenance.

LOUGHNAN and BAYLISS [1961] described a large deposit of bauxite near Weipa, Queensland, Australia which formed from quartzose rocks containing less than 4 percent alumina.

The fire clay and the bauxites of Muzaffarabad and Kotli Thesil do not show any degree of positive correspondence with the immediate underlying precipitation breccia of Muzaffarabad Formation, as suggested by LOUGHNAN and BAYLISS for the origin of Weipa bauxite of Queensland from quartzose rocks.

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THE MINNICHHOF METEORITE

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ABSTRACT

The dominant minerals of the Minnichhof meteorite are olivine ($Fa_{25.3}$) and hypersthene ($Fa_{21.5}$) with nickel-iron (kamacite, taenite, plessite), plagioclase (An_{12}), diopside of minor quantity and accessory chromite, merrillite, ilmenite, daubréelite and limonite. Specific gravity: 3.53.

On the basis of the given electron-probe microanalysis as well as the complete chemical analysis, *X*-ray diffractometric diagrams, spectrographical and mineral-petrographical examinations the stone is an olivine-hypersthene chondrite [MASON, 1962 and 1965], equilibrated, ordinary chondrite [DODD—VAN SCHMUS—WOOD, 1967] which belongs to the L-group [UREY—CRAIG, 1953] and can be classed into the L5—L6 types [VAN SCHMUS—WOOD, 1967].

INTRODUCTION

The meteorite fell on May 27, 1905, 10^{45} a.m. on the territory of the village Malomháza (Felsőpulya district, Sopron county, Hungary). Its geographical coordinates: 47° 32′ N and 16° 38′ E. In the report, which can be considered as a source, F. BERWERTH [1912] applies the German — Minnichhof — name of this locality, and the Hungarian name was misdescribed as a synonym.

Though in the L. TOKODY–M. DUDICH VENDL catalogue [1951] Malomháza is denoted as the locality of fall, both the PRIOR–HEY [1953] and HEY [1966] catalogues use Minnichhof. Because the village was annexed in 1921 to Austria, and because till that date neither the detailed study of the stone, nor the elucidation of the name of the locality were performed, we also use and accept the name Minnichhof (Burgenland, Austria) in our catalogue [1961] and the present paper as well. We know a little about the story of the stone and about the circumstances and the date of acquisition, because — owing to the 1956 fire in the Mineralogical and Petrographical Depertment of the Hungarian Natural History Museum — the inventory books were destroyed. It was identified on the basis of the inventory number — being on the stone — and of the data included in the 1951 catalogue. The weight of the present piece proved 54.8 g. less than the value given in the TOKODY–DUDICH catalogue, at the same time a thin, wedged slice was missing. Its volume is proportional to the deviation in weight. The missing slice has probably been cut for the purpose of a designed investigation, but no evidence can be found referring either to the examination or to its results. This fact made necessary the more thorough investigations of the meteorite, which has been considered only as a white chondrite. Sampling and testing could have been carried available for the most important partial experiments.

EXPERIMENTAL METHODS AND TECHNIQUE

Sampling

The material to be tested was obtained by cutting slices parallel with the previously cut surface. One slice was used for making thin section and gave the dominant part of the tested material, the other one served as preparatum for the electron-probe microanalysis (at the same for ore microscopic examination) and another thin section and the remaining parts supplied the tested material as well. After making the electron-probe preparation and thin section, about 2 g of the particles (2–3 mm in diameter) was reserved for the determination of specific gravity. The residue (about 20 g) was pulverized graduelly to 0.2 mm ∅ grains, while paramagnetic fraction was separated by a handmagnet and the pieces of fusion crust were also selected. The further separation into silica and non-silica mineral fractions was carried out by centrifuge on Clerici-Solution of 4.22 spec. gravity. The result of the separation to heavy and light fractions was controlled under microscope, and the procedure was repeated till the mixing of the two fractions diminished to the minimum. Thereafter both fractions were centrifuged in distilled water till thallium-ionfree state. It was controlled with 0.1 n solution of KJ. The material of both heavy and light fractions were desiccated in water bath and subjected to trace element detection, X-ray examination and chemical analysis.

Electron-probe microanalysis

The quantitative chemical analyses of each mineral were carried ot by JEOL JXA–5 electron-probe microanalyser in the Geochemical Research Laboratory of the Hungarian Academy of Sciences. Accelerating voltage: 25 kV. The polished section and the standards were coated by aluminium simultaneously. The applied standards for Fe (in silicate phase) Mg and Si — olivine, for Al — topaz, for Na — oligoclase, for K — orthoclase, for Fe — iron metal, for Ni — nickel metal, for Ti — titanium metal. For correction computer was applied [cp. G. NAGY, 1970].

X-ray analysis

Two X-ray powder diffractograms were made both from the heavy and the light fractions on a Philips–Mueller 111 type X-ray diffractometer, using Cu radiation and Ni filter, at 26 kV, 36 mA, goniometer speed: 2°/min and 1°/min, intensity factor: 8, slits: 1°, 0.1°, registration speed: 27 mm/min, angular range: 2°–63° 2Θ. (Analyst: MISS A. SZEMETHY.)

Spectrophotometric analysis

Sample preparation: grain size 60 μ, mixed with Pd containing spec. pure carbon powder in equal proportion. — Electrode: 6 mm ∅ carbon rod — Excitation: A. C. arc, 16 amper, analytical gap: 3 mm — Exposure time: 180 sec till the total evaporation — Optic: Jobin–Yvon Z3 Quartz Spectrograph, two-step filter; — Slit width: 15 μ; — Evaluation: Zeiss Schnellphotometer, 1-transformation, background correction, Pd internal standard.

In the heavy fraction the values of Pd and Zr were not analyzed because of the interfering effects of Fe and Co. (Analyst: MRS. M. VIGH).

Chemical analysis

After decomposing by aqua regia the material of the heavy (2 g) and the light (14 g) fractions, the solution was repeatedly evaporated by 1:1 diluted HCl solution in order to eliminate nitrates. Finally the dry residue was dissolved in hot 1:10 diluted HCl solution and filtered. The residue was repeatedly washed till chlorid-free, the iltrate was reserved. After burning, the insoluble residue was fused by Na_2CO_3, issolved in HCl and combined with the reserved filtrate, and evaporated to dryness. his was followed by the usual silicate analytical method. Silica was determined by ravimetric method. Total iron, aluminium, manganese, phosphorus, chrome and cobalt were determined by spectrophotometric method. The gravimetric determination of nickel was made after GROVES' [1951] method, the determination of metallic ron after the WILNER–MERCK method [Merck, 1902] and that of the alkalies was carried out by flamephotometry. The sulphur content of troilite was determined fter RADER–GRIMALDI's [1961] gas-volumetric method.

The reserved material for specific gravity determination was weighed, put in Cl_4 into a picnometer, was boiled in vacuum, induced by pneumatic water-pump: or 3 minutes. The spec. gravity was determined at 20° C.

MEGASCOPIC DESCRIPTION

The shape of the meteorite can be compared to a scalene pyramid of blunted dges and corners. Its frontside is rounded, the rearside is almost quite flat, the ross-section is trapezoid, of the four further planes two are convex and two are oncave *(Fig. 1/a)*. Its greatest diamaters are: 71.52 and 45 mm; weight: 462.2 g. n the sides of the stone two bigger and several smaller piezoglypts can be seen. riginally the whole stone was covered by a black — in thin veins brownish-black — usion crust. This crust of velvety lustre is relatively uniform in thickness, on an verage of 0.5 mm, growing thin in some places. 1–2 mm thick reddish dark-brown oloured oxidized crust can be found sporadically, terminated vaguely towards the nterior. The surface of the stone is ragged due to the bulging chondrules and metallic articles, i.e. to the irregularly scattered furrows and flutings, in a lesser extent. n the bulging structural elements the fusion crust has a weak vitreous lustre. On he crust of the meteorite there are some smaller cracks, while inside one dominant nd several very thin ones *(Fig. 1/b)*. The specific gravity of the meteorite is 3.52.

The bulk of the stone is a dark-grey coloured, compact material of medium ardness. In this material relatively few chondrules and irregularly disseminated etallic particles are embedded. Chondrules break together with the groundmass. One part of them is white, round, generally 1.0 mm (0.5–1.4 mm) in diameter, the thers are grey with ovoid, round or irregular cross-section, mostly 1.5 mm (1.0–.0 mm) in diameter. Some of the grey chondrules show spherulitic texture. The etallic particles on the fresh cut surface are of vivid metallic lustre, generally 0.2 mm (0.1–1.2 mm) in diameter, often jagged along the irregular contours.

MINERALOGICAL COMPOSITION AND STRUCTURE

The dominant minerals of the stone are: olivine and hypersthene, a relatively igher amount of palgioclase and diopside, Ni-Fe particles and troilite. Chromite, errillite, ilmenite and daubréellite, are accessories as well as the secondary limonite Tables 1, 2, 3).

Fig. 1.
a) Photograph of the Minnichhof meteorite. *b)* Photograph of an other side of the stone.

Crystallized groundmass intergrown with the condrules is characteristic of the texture. There are only a few sharply defined chondrules, glass is completely missing. Metallic components are irregularly scattered in the groundmass, they are frequent in schondrules and on the outer edge of them, and appear in narrow veinlets as filling metarial as well.

The *olivine* ($Fo_{75}Fa_{25}$) crystals are colourless, some of them have light-brown, reddish-brown colour. Colour can be observed along the cleavages and parting traces and in irregularly scattered small patches. Most of the crystals are allotriomorphic, wavy margins are frequent. Hypidiomorphic or almost idiomorphic, dipyramid-terminated crystals of short prismatic habit are uncommon. Its biggest dimensions (600–800 μ) appear in the monosomatic chondrules. In the porphyritic chondrules this value is 500–600 μ, in the matrix 50–500 μ. It contains plagioclase, monoclinic and orthorhombic pyroxenes, troilite and Ni-Fe particles as inclusions *(Fig. 2/a, b)*.

On the basis of electron-probe microanalyses perfected on three crystals the composition of olivine is uniformly $Fe_2SiO_3 \sim 25.3$ mole% (Tables 4, 5). On the X-ray diffractometer chart of the light fraction, the d_{130}-reflection shows sharp and well-defined peaks; on the basis of YODER–SAHAMA [1957] determinative curve this corresponds to $Fe_2SiO_4 = 25$ mole%. According to POLLACK's and CHI's method [1972] we got a value slightly lower than the former ones.

Hypersthene ($En_{76}Fs_{22}Wo_2$). The dominant part of the crystals are water clear and pale pinkish, some of them are cloudy having a faint shade of greyish colour. Most of them are allotriomorphic, but in the chondrules some hypidiomorphic

TABLE 1

Results of chemical and trace analyses

Chemical composition (in weight %)				Trace elements (in ppm)		
	bulk analysis	heavy fraction	light fraction		heavy fraction	light fraction
SiO_2	40.44	1.09	46.06	P	nd	1200
TiO_2	0.18	0.21	0.18	Ti	350	650
Al_2O_3	2.12	0.25	2.41	Cr	nd	2000
Fe_2O_3	0.18	0.02	0.20	Mn	800	2000
FeO	14.56	1.54	16.39	Co	1000	35
MnO	0.34	0.06	0.38	Ni	nd	1250
MgO	25.53	1.00	29.04	Cu	75	35
CaO	2.44	0.17	2.76	Ge	8	<1
Na_2O	1.04	0.38	1.14	Zr	nd	200
K_2O	0.12	0.10	0.12	Ga	5	1
P_2O_5	0.18	0.02	0.21	Mo	8	<2
Cr_2O_3	0.48	1.96	0.26	Sn	<1	<1
$-H_2O$	0.02	—	0.02	Pb	<2	<2
Fe	6.84	53.33	0.21	V	7	nd
Ni	1.24	8.24	0.24	Zn	220	nd
Co	0.02	0.17	0.004	Ag	3.4	nd
Fe	2.87	19.81	0.45	Bi	<3	nd
S	1.64	11.38	0.26			
S	0.05	0.46	—			
Total	100.29	100.19	100.334			

TABLE 2

Results of electron-probe analyses (in wt.%)

Metal phase	Fe	Ni	S	Total	Analysed particle
Kamacite	91.5	6.1	—	97.6	Main mass *(Fig. 6.)*
Taenite	82.5	15.9	—	98.4	Inclusion *(Fig. 6a, b)*
Taenite	—	40.3	—	—	Diffusion border of the inclusion *(Fig. 6c)*
Troilite	63.1	—	35.9	99.0	Average value of three particles (two of them on *Fig. 7a, b*)

Silicate minerals	SiO_2	TiO_2	Al_2O_3	FeO	MgO	CaO	K_2O	Na_2O	Total
Olivine	37.5	—	—	23.7	39.2	—	—	—	100.4
Hyperstehene	55.1	—	—	14.2	28.1	1.0	—	—	98.4
Diopside	55.0	0.2	0.1	4.2	16.0	23.7	—	—	99.2
Oligoclase	64,6	—	21.6	—	—	2.4	0.8	10.5	99.9

elongated prismatic crystals, and in a monosomatic chondrule an equidimensional, 800 μ large and almost idiomorphic crystal can be observed. Except some twinned specimens they are monocrystals, 50–60 μ of grain size. Hypersthene occurs mainly

TABLE 3

Mineral composition, calculated from values of chemical and electron-probe analyses (in wt.%)

Or ($K_2O.Al_2O_3.6\,SiO_2$)	0.70		
Ab ($Na_2O.Al_2O_3.6\,SiO_2$)	8.12		
An ($CaO.Al_2O_3.2\,SiO_2$)	1.18		
Feldspar $Or_4Ab_{84}An_{12}$		10.00	
En ($MgO.SiO_2$)	14.25		
Fs ($FeO.SiO_2$)	5.32		
Wo ($CaO.SiO_2$)	0.41		
Orthopyroxene $En_{76}Fs_{22}Wo_2$		19.98	
En ($MgO.SiO_2$)	4.45		
Fs ($FeO.SiO_2$)	0.47		
Wo ($CaO.SiO_2$)	3.93		
Ilm ($FeO.TiO_2$)	0.04		
Clinopyroxene $En_{45}Fs_7Wo_{48}$		8.89	
Fo ($2\,MgO.SiO_2$)	31.39		
Fa ($2\,FeO.SiO_2$)	15.40		
Olivine $Fo_{75}Fa_{25}$		46.79	
Silicate minerals			**85.66**
Merrillite ($3\,CaO.Na_2O.P_2O_5$)	0.48		
Ilmenite ($FeO.TiO_3$)	0.30		
Chromite ($FeO.Cr_2O_3$)	0.64		
Limonite (FeOOH)	0.20		
Accessory Minerals			**1.62**
Ni-Fe ($Ni_{15.4}Fe_{84.4}Co_{0.25}$)	8.08		
Troilite (FeS)	4.51		
Daubréelite ($FeCr_2S_4$)	0.11		
Metal phase			**12.70**
Silicate + accessory minerals: 87.28			
Metal + sulphide minerals: 12.70			
Total 99.98			

in chondrules, in porphyritic and barred form, or in groups of oriented intergrown crystal groundmass it occurs as phenocryst or in interstitial position. As inclusions diopside and feldspar are most common.

On the basis of electron-probe microanalysis (Tables 4, 5) the composition of hypersthene can be characterized by the value $FeSiO_3 \sim 21.5$ mole%. Though the calculated atomic ratios — when compared to the data known from the literature [DEER, HOWIE, ZUSSMAN, 1965] are referring to a composition similar to the bronzite, the reflections on the X-ray pattern correspond to hypersthene and not to bronzite. Based on electron-probe microanalysis the monoclinic pyroxene is *diopside* ($En_{45}Fs_7Wo_{48}$), its calculated amount is 9 wt%. On X-ray diagram it is represented by well-defined reflections. Under the microscope it is colourless and transparent with characteristic cleavage traces. Extinction angle: 38–42°. It occurs mainly in the crystalline groundmass *(Figs. 2/c, d)*, less in the chondrules, and can be found as inclusion in olivine and hypersthene. The dimension of dominantly allotrio-

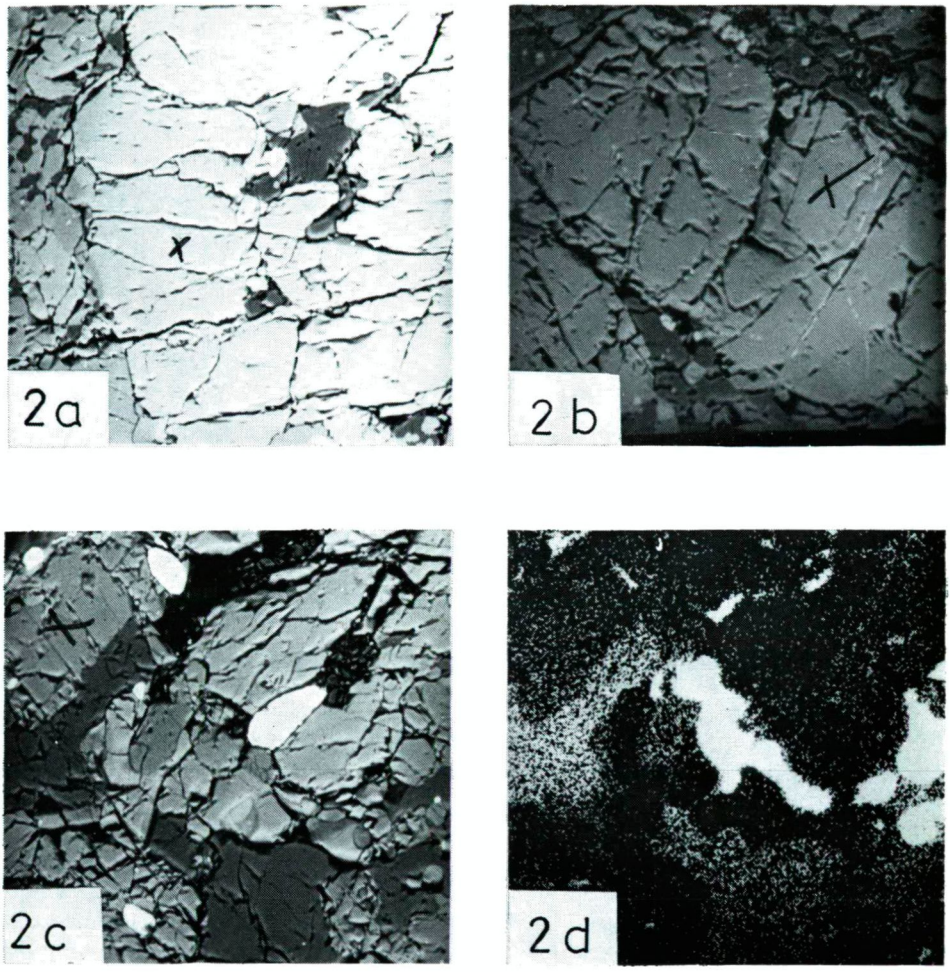


Fig. 2.

a) Back-scattered electron picture of olivine, with plagioclase (gray), Ni-Fe (white) inclusions in it. 25 kV, 300x.

b) Back-scattered electron picture of a hypidimorphic olivine crystal. 25 kV, 300x.

c) Back-scattered electron picture of hypersthene (light gray), diopside (gray in center and right), and interstitial plagioclase (dark gray). 25 kV, 300x.

d) CaKα x-ray picture of 2c. Hypersthene (dark area), diopside (white), plagioclase (light). 25 kV, 300x.

morphic — rarely hypidiomorphic-crystals in the groundmass is 60–80 μ, infrequently 100–200 μ. In sections its 200–300 μ large crystals form discontinuous ring around the porphyric chondrules. In the radial chondrules it can be observed alternating with orthopyroxene and plagioclase in columnar or fibrous development.

The amount of feldspars is 10 wt%. On the basis of the electron-probe microanalysis the modal composition is $Or_4Ab_{84}An_{12}$ mole%, i.e. *plagioclase,* acid oligoclase. On the X-ray diagram the reflections of plagioclase give characteristic sharp peaks corresponding to the high oligoclase [GOODYEAR, DUFFIN 1965]. In polarized

TABLE 4

Crystallochemical data, calculated from the results of electron-probe analysis

Mineral	Composition in wt. %		Numbers of ions on the basis of		Atomic ratios	Formula
Olivine			4 oxygens			$Mg_{1.49}Fe_{0.51}[Si_{0.96}O_4]$
	SiO_2	37.5	Si	0.956		
	FeO	23.7	Fe^{2+}	0.505 } 1.99	25.3	
	MgO	39.2	Mg	1.489 }	74.7	
	Total	100.4				
Hypersthene			6 oxygens			$(Mg_{1.52}Fe_{0.43}Ca_{0.04})\,[Si_2O_6]$
	SiO_2	55.1	Si	2.002		
	FeO	14.2	Fe^{2+}	0.432 }	21.7	
	MgO	28.1	Mg	1.522 } 1.99	76.4	
	CaO	1.0	Ca	0.039 }	1.9	
	Total	98.4				
Diopside			6 oxygens			$Ca^{[8]}_{0.93}(Mg_{0.88}Fe_{0.13}Al^{[6]}_{0.004}Ti_{0.006})\,[Si_2O_6]$
	SiO_2	55.0	Si	2.020		
	TiO_2	0.2	Ti	0.006 }		
	Al_2O_3	0.1	Al	0.004 }		
	FeO	4.2	Fe^{2+}	0.129 } 1.95	6.6	
	MgO	16.0	Mg	0.876 }	45.2	
	CaO	23.7	Ca	0.933 }	48.1	
	Total	99.2				

TABLE 5

The more important chemical parameters

	wt. %		wt. %
$Fe_{metallic}$	= 6.82	$Fe_{metallic}/Fe_{total}$	= 0.45
$Fe_{sulphidic}$	= 2.89	$Fe_{metallic}/Ni$	= 5.5
$Fe_{metal\ phase}$	= 9.71	Fe_{total}/Ni	= 17.0
$Fe_{silicate\ phase}$	= 11.45	Fe_{total}/SiO_2	= 0.52
Fe_{total}	= 21.16	SiO_2/MgO	= 1.58

Fe_2SiO_4 in olivine = 25.3 mole %

$Fe^{\circ}_{olivine}$ = 18.4 wt. %

$FeSiO_3$ in hypersthene = 21.5 mole %

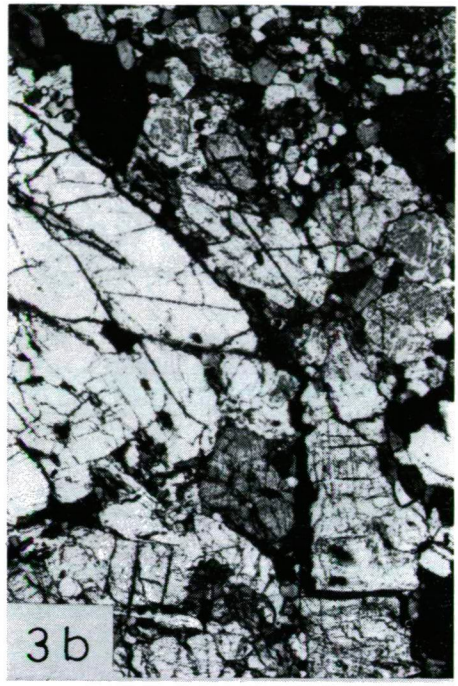
$Fe^{\circ}_{hypersthene}$ = 11.0 wt. %

light the allotriomorphic crystals are quite clear showing no cleavages, $n \leqq n_{Canada\ balsam}$. The crystals are 20–120 μ large, finer grains form aggregates with pyroxene and olivine in some patches. The rarely observable, weak, uneven extinction marks the recrystallization of glass. One third of the crystals are twin-lamellar according to the albite law. The dominant part of plagioclases is in interstitial settlement, related both to the groundmass and to the chondrules *(Figs. 2/c, d* and *3/b, c, d)*. In accordance with the texture of chondrules plagioclase appears as a component of the matrix in the barred type, while it is mosaic-like, interstitial in the radial chondrules and forms inclusions in the monosomatic orthopyroxene ones. Within a little patch of the thin section, in the plagioclase aggregates, fine grained orthopyroxene crystals are embedded.

Regarding their shape, texture and mineralogical composition the *chondrules* are rather varied, at the same time they are quite uniform in dimensions (1–2 mm). Most of the chondrules have round or oval cross-section. In the microscope they cannot — or can hardly — be distinguished from the groundmass. More exactly: while the cores of the monosomatic, radiated and barred chondrules are clearly discernible the polysomatic, mixed and porphyritic chondrules merge absolutely into their well-crystallized environment *(Fig. 3/a)*. The monomineralic chondrules are often surrounded by loose or ragged rim of differing minerals, or metallic particles, troilite. These rims indicate the original contour of the condrule, therefore from the aspect of rock-texture these chondrules can be regarded as transitional members *(Figs. 3/b, c, d)*. The few, sharply delineated chondrules are whole and have round cross-sections, or are fragments. The former ones are of monosomatic or excentro-radial texture. Some of the broken chondrules are 20–80 μ diameter and are localized generally along fissures filled by troilite.

Regarding the composition, the chondrules broken by micro-faults are: monosomatic orthopyroxene chondrule with plagioclase inclusions of undulatory extinction *(Figs. 4/a, b)*; polysomatic orthopyroxene chondrule; barred olivine and porphyric (mixed) orthopyroxene-olivine chondrule. The other part of broken chondrules, restricted to some pieces, are embedded into the crystalline groundmass, they are isolated or joined with other chondrules and consist of fibrous and barred ortho-and monoclinic pyroxene *(Figs. 4/c, d)*.


3a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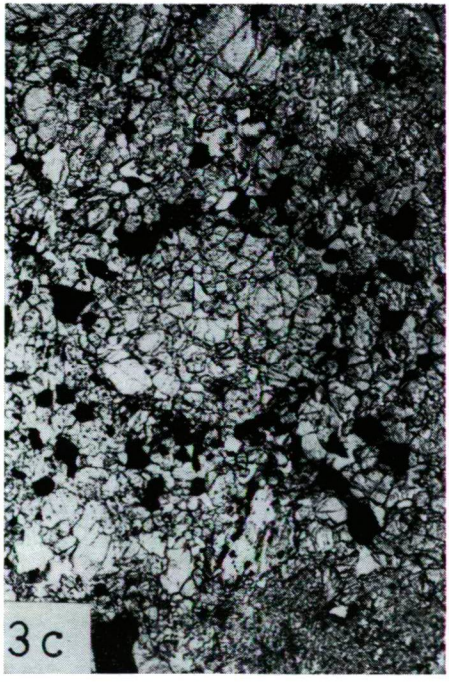
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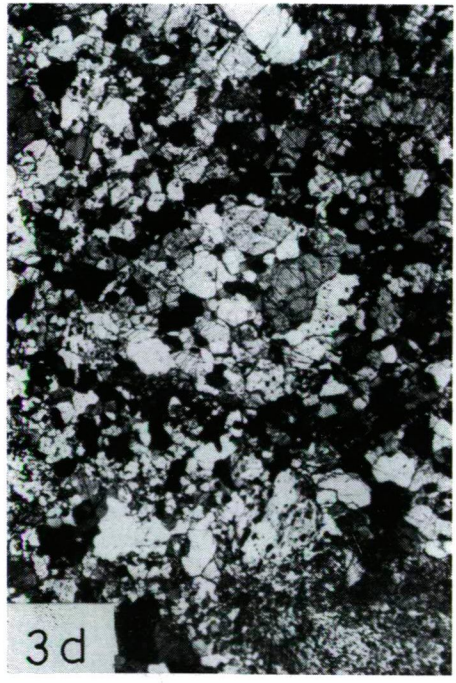
3d

Regarding their mineral composition the chondrules mainly consist of the dominant minerals. Porphyric olivine is more common than the barred one, sometimes monosomatic. Orthopyroxene is abundant in monosomatic and porphyric chondrule, it is rare in polysomatic barred chondrule intergrown with diopside, and there are only a few specimens of structurless appearance. In the latter group, among the 50–200 μ large allotriomorphic hypersthene crystals, intersticial diopsides of 15–25 μ grain size can be observed. As condrule-forming mineral the diopside as a fibrous and radial aggregate is rarely observable.

In addition to the above discussed role of the plagioclase it must be noted that plagioclase — similarly to the metallic particles — can be found as intersticial component in each chondrules or surrounded them as aggregates *(Figs. 3, 4, 5)*.

The grain size of the microholocrystalline *groundmass* is varying. The patches consisting of 15–50 μ large ortho-, clino-pyroxene and plagioclase crystals can be considered as typical matrix, still they constitute only a small part of the groundmass. On the contrary the microcrystalline groundmass of porphyric texture is widely distributed. It consists of 20–75 μ large allotriomorphic ortho-, clinopyroxene, olivine and plagioclase crystals 150–200 μ large allotriomorphic — rarely hypidiomorphic — olivine and some orthopyroxene crystals. The latter are embedded into the former ones. The scattered or interstitial 40–100 μ large metallic particles and troilite crystals bordered by irregular contoures must also be considered to belong to the groundmass.

Metallic particles

On the basis of the optical investigations, chemical analyses and X-ray diagrams of the heavy fraction it can be stated that the metallic phase is represented by Ni-Fe particles, troilite, minimal amount of chromite, ilmenite, daubréelite and secondary limonite (Tables 1, 2, 3).

The shape and composition of Ni-Fe particles are very variegated. Two groups can be quite clear distinguished. The equidimensional, rounded, drop-like 60–200 μ large particles can be put to the first group and the elongated 400–1200 μ large particles showing jagged borders into the second one. One part of the metallic particles are broken and some of them from loose chain along the cracks which is observable even macroscopically.

Kamacite of minor amount occurs rarely as isolated grains, most of it is joining with other Ni-Fe particles and with troilite. Single crystals or polycrystalline masses are common, hypidiomorphic forms are exceptional. Well-developed, straight, broad NEUMANN-lines forming at most four-directional network can be found only in the larger crystalline aggregates. On an other particle two types of texture can be observed: the first shows close parallel straight lines, the other faintly distorted ones. Finally, between two kamacite particles, like a bridge, a third kamacite particle can be found patterned by narrow lines. Taenite borders are very common and

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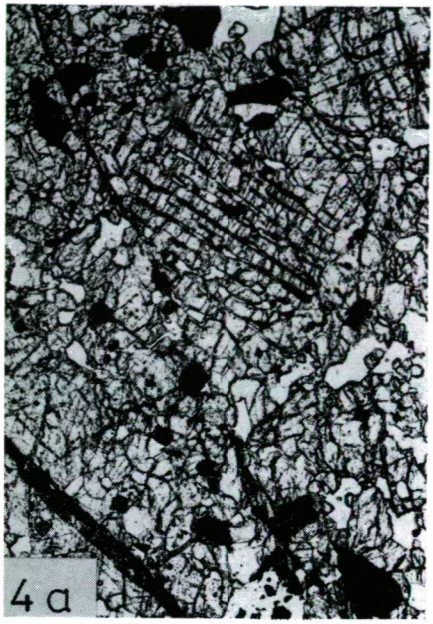
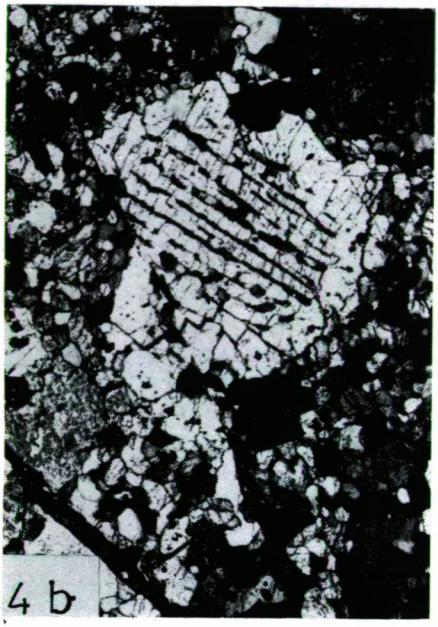
Fig. 3.

a) Textural picture. The chondrules merge into the crystalline groundmass. +N, 27.5x.
b) Monosomatic, barred hypersthene chondrule (white) surrounded by clinopyroxene and plagioclase. +N, 68x.
c) Crystalline porphyric chondrule (centre) in troilite and chromite (black) internal ring, bordered by plagioclase, diopside, troilite (white, grey, black) in the crystalline groundmass. //N, 27.5x.
d) id. +N.


4 a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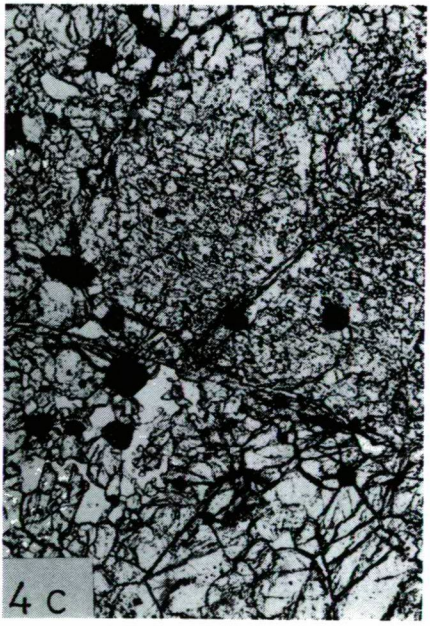
4 c


4 d

taenite inclusions and intrusions as well *(Fig. 6)*. In reflected light kamacite is greyish white with strong metallic lustre. After etching by "nital" it turns to light-grey, reflects weaker. On the basis of electron-probe microanalysis its Ni-content is 6.1 wt%.

Taenite — in addition to the above mentioned appearance as diffusion border — can be observed as discrete particle, too. In reflected light it has a yellowish-white colour and strong metallic lustre, which characteristics are preserved after the etching as well. Its Ni-content is 16–30 wt%, though very fluctuating (max. 40 wt%) even with in a certain particle *(Fig. 6)*. It is attached most frequently to kamacite and to plessite; the phase boundaries can be either straight edges or irregularly jagged contacts. It generally contains plessite as inclusion.

The alternately fine or coarse grained *plessite* is the most common metallic component. The size of grains for sure identification under microscope are 0.5 mm. The particles are elongated, dissected by appendices and embayments. The vermicular intergrowth of taenite and kamacite appears mainly after etching, and this characteristic structure can be studied on the screen of microanalyser as well *(Figs. 7/c, d)*. The coarse plessite particles have generally broad Ni-diffusion borders. In the particles bearing teanite or kamacite cores, the fine plessite forms rim, sometimes little droplets, within the diffusion border. WIDMANSTÄTTEN structure could have been observed only on a single particle found on the one side of a larger, elongated, mainly polycrystalline kamacite particle, varying in composition. It is limited by a faint line towards the interior of the kamacite, and by a straight fracture along the margin of the particle. On the other side of the micro-fracture a teanite splinter can be observed. In the inner part, of the splinter of Ni-diffusion border fine plessite rim is to be found in one part some tiny fine plessite droplets are observable in the other.

The *troilite* forms allotriomorphic crystals with irregular contours, and rarely hypidiomorphic ones with hexagonal cross-section. These appear as single crystals or form irregularly distributed aggregates. Troilite can be observed as inclusion in silicates, as rim around some chondrules and as filling mineral in veinlets. Its amount is 4.5 wt%, on the basis of electron-probe microanalysis its composition is: Fe$\sim$50.2 and S$\sim$49.8 mole%. The dimension of troilite drops is generally 150–200 μ, the diameter of the amoebiform aggregates is 400–600 μ long. The troilite is light-yellow, the strongly reflecting patches are fresh-yellow, with a characteristic bronze tinge. Most of the crystals are cracked, dissected by cleavage and parting traces and have rugged rims *(Figs. 7/a, b)*. In some cases pressure effect, polysynthetic twinning can be seen. It rarely joins with Ni-Fe particles, but when it does, it can be found intruding into them and contacting the silicate-surrounding only with one side. In other cases troilite can be seen in the embayments of metallic particles or joining

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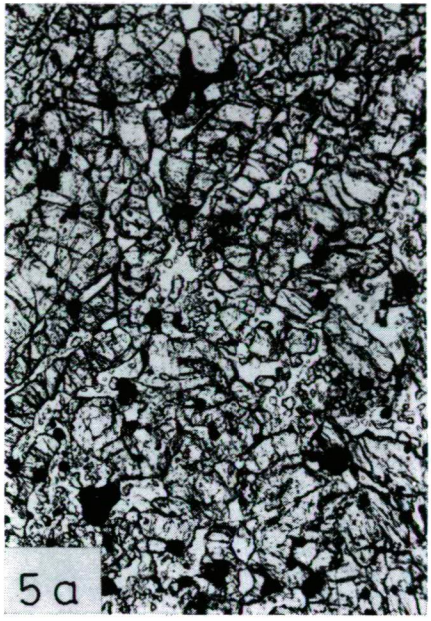
Fig. 4.

a) Disrupted and dislocated monosomatic hypersthene chondrule (upper third) with interstitial plagioclase (white), ore and troilite inclusions (black), troilite veins (black NW—SE oriented) //N, 68x.

b) id. +N. Note: along the cleavage oriented NW—SE the hypersthene chondrule (white) moved by 226μ.

c) Radial, fibrous pyroxene chondrule sharply delineated by two fractures (above right) in crystalline groundmass, with a lot of interstitial plagioclase (white), and scattered metallic particles (black). //N, 68x.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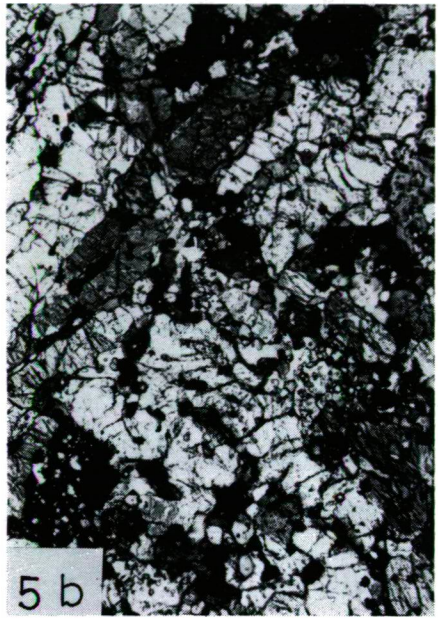
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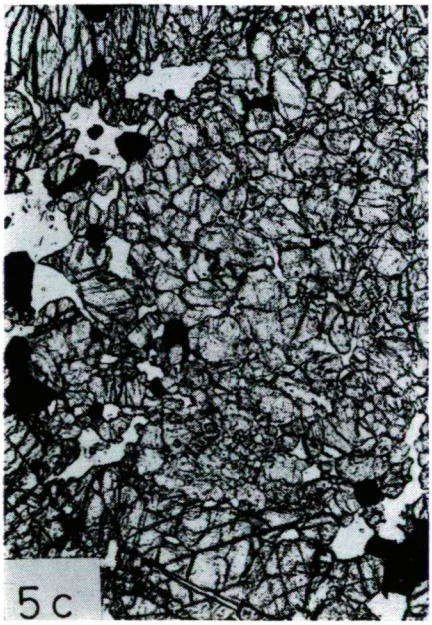
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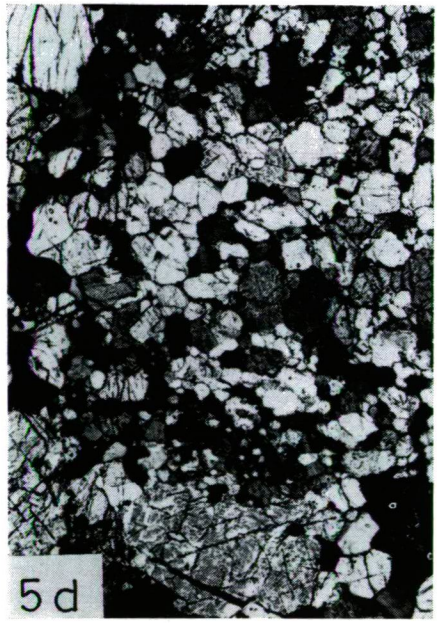
5 d

them as tags, finally forming bridge between two Ni-Fe particles. The irregular aggregates have a pore-filling role, the equant particles are embedded into the crystalline mass of silica minerals as interstitial material.

Accessory minerals

Chromite occurs as some larger (200–400 μ) hypidiomorphic and as some smaller (15–20 μ) allotriomorphic crystals. On the larger crystals octahedric parting can be observable, on the rim of a rhombohedric crystal in cross-section a slight alteration can be seen. Fine grained chromite in regular form can be examined only in a single place in the preparatum, in a chondrule, where its grains form a ring parallel with the margin of the chondrule *(Figs. 3/c, d)*. *Ilmenite* succeeded to be identified only in a single case. In a red limonitic environment, in the vicinity of a Ni-Fe particle it forms a group consisting of three plates of 20–25 μ. The hypidiomorphic, tabular crystals are joined each other by crystal faces. On all the three crystals lamellar twinning ($10\bar{1}1$) can be recognized.

Daubréelite could be identified only on X-ray diagrams and in spite of the thorough investigation of troilite particles they could not be observed under the ore microscope. The small dimensions of the preparatum may be responsible for it. After the normative calculations we established the amount of *merrillite* in 0.48 wt%. Its allotriomorphic crystals occur sporadically in the groundmass, in thin sections they can be hardly observed.

Secondary *limonite* appears as filling material along the "in situ" disruptions or along the rim of metallic particles, troilite and olivine. The thickening of veins results in patches. Limonite forms thin coating on the oxidized parts of the fusion crust.

INTERPRETATION OF OBSERVATIONS

Owing to the fact, that the purpose of the present paper was — first of all — the thorough testing of the Minnichhof meteorite and consequently the publication of the results arranged in certain respect, therefore, only the measured and observed data most important from the viewpoint of systematization are stressed. Similarly, we tried to discuss our conclusions relating to genetics within a compass corresponding to the profoundity of investigations. The material of Minnichhof meteorite after MASON's system [1962, 1965] is an olivine-hypersthene chondrite. According to the ROSE–TSCHERMAK–BREZINA [1812, 1872, 1904] system it belongs to the intermedier crystalline chondrite group, after the PRIOR-system [1920] it can be ranged into the hypersthene-olivine chondrite group. On the basis the data of bulk analysis, the electron-probe microanalysis of the dominant minerals and on the strength of the index-numbers formed of these values the meteorite belongs to UREY–CRAIG's [1953] L-group. Comparing the above data to the mineral constituents and textural

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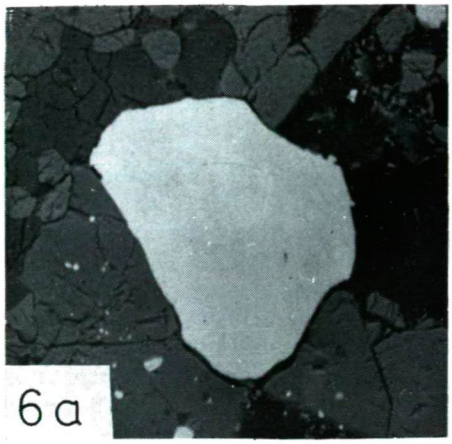
Fig. 5.

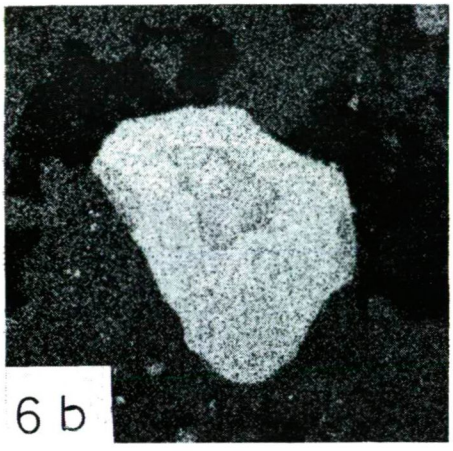
a) Pyroxene and olivine bars, interstitial plagioclase (white) and metallic particles (black) in mixed, barred chondrule //N, 68x.

b) id. +N.

c) Fine grained mixed chondrule, interstitial plagioclase (white) — inside and around the chondrule — metallic particles and troilite (black) in holocrystalline porphyric groundmass. //N, 68x.

d) id. +N.





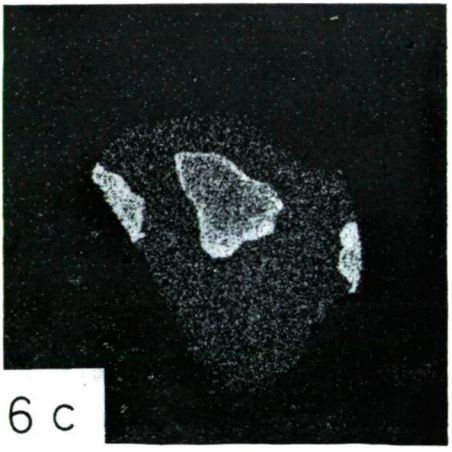


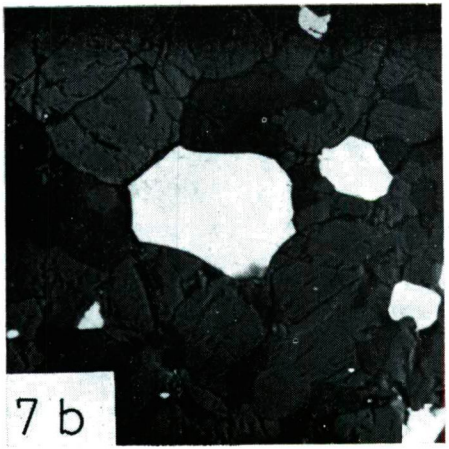
Fig. 6.

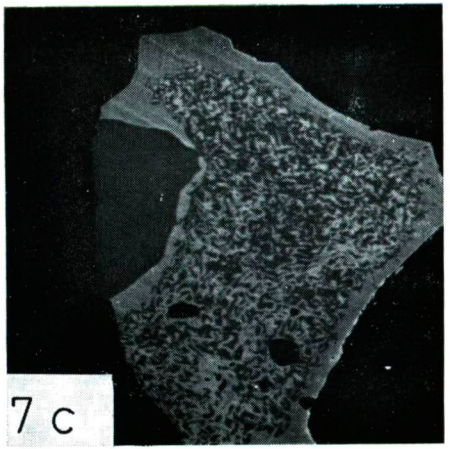
a) Back-scattered electron picture of kamacite particle with taenite inclusion and intrusions, surrounded by silicates. 25 kV, 300x.
b) FeKα x-ray picture of 6a, 300x.
c) NiKα x-ray picture of 6a, 300x.

Fig. 7.

a) Back-scattered electron picture of polycrystalline troilite intruded into silicates. 25 kV, 300x.
b) Back-scattered electron picture of troilite embedded in silicates. Some of the crystalls are nearly equant. 25 kV, 300x.
c) Back-scattered electron picture of coarse plessite with irregular taenite border. 25 kV, 300x.
d) Back-scattered electron picture of coarse plessite. Same as 7c, enlarged. The line indicates the track of the distribution profiles in 7e and 7f. 25 kV, 1200x.
e) FeKα distribution along the line of 7d, 1200x.
f) NiKα distribution along the line of 7d, 1200x.


7a


7b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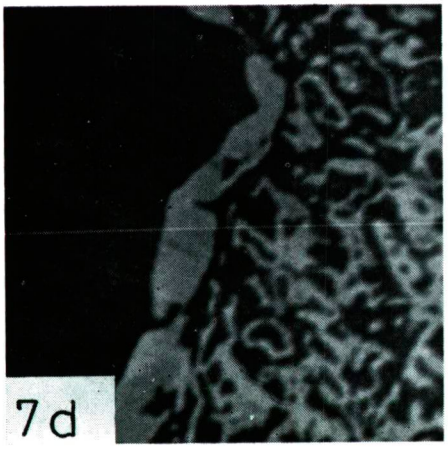
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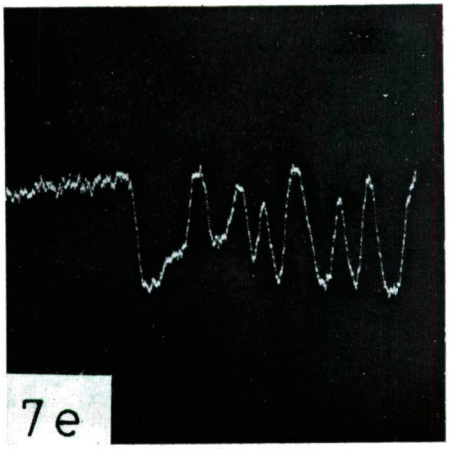
7e


7f

characteristics, is near to the L5–L6 types of VAN SCHMUS–WOOD's [1967] chemical-petrological classification. Finally, considering the DODD–VAN SCHMUS–KOFFMAN [1967] system the meteorite is an equilibrated ordinary chondrite with an advanced textural integration. Comparing the values of chemical analysis, modal and normative mineralogical composition to the data published by MASON [1965]:

(i) they show concordance: FeO, MgO, Cr_2O_3, TiO_2, Fe, Ni wt% in chemical analyses, and Ol, Or, Ab wt% values in normative mineralogical composition;
(ii) they are similar: Al_2O_3, Na_2O, K_2O, Co, total silicate, total Fe, i.e. Hy, Cm, Ilm wt%;
(iii) or they are within the given limits: SiO_2, CaO, P_2O_5, FeS, i.e. Di, An wt%.

The ranging into the olivine-hypersthene group is supported by the values Fa=25.3 mole% in olivine and Fs=21.5 mole% in orthopyroxene which values were verified by different investigations. If we compare the data of the 94 superior analyses published in UREY–CRAIG's paper [1953] to the results of our bulk analysis we get a very similar result. In some values of the silicate phase the difference is 1–0.5 wt% in SiO_2, MgO and CaO, — similarly to the MASON-values — but this difference is balanced in the total sum of the silicate phase. Greater difference appears at the metallic phase and the FeS values. More exactly: though the total amount of metallic Fe, Ni and Co is almost equal with the average, because of the relatively less amount of Fe(+Co) and Fe-sulphides and of the relatively greater amount of Ni, the values of the Fe-metallic phase, Fe total and Fe_{metal}/Ni shows a deficit of 1%. This means that it draws near to LL-group [KEIL and FREDERIKKSON, 1964], at the same time the value of Fe_{total}/Ni is characteristic to the H-group (Table 5). The above discussed quantitative relationships cause that in the wt% relation of silicate: troilite: iron phase the sulphide iron: iron ration corresponds to the value 85:6:9 given by UREY [1952], but the wt% relation of silicate: metallic phase does not correspond to it. Concerning the GOLDSCHMIDT-values (10:1:2), the discussed ratios do not answer to them at all. In conformity with the above mentioned aggreements and similarities can be pointed out with WIIK's analytical values [1956]. If we compare our data to the values of the table in WAHL's paper [1950] which contains the average, normative mineralogical composition of L-group, in case of Fo, Fa, Cm and Ilm we get absolute concordance, the values of other minerals are more or less different.

The results published in the chapter on chemical differences of the paper written by VAN SCHMUS and WOOD [1967] and the indices gained by the new grouping of chemical parameters confirm that the tested meteorite belongs to the L-group. In accordance with the above details of this paper we can establish that the homogeneity of olivine and pyroxene, the lack of the low Ca-clinopyroxene and the glass, the composition, grain size and the arrangement of plagioclase; further the sort, shape, Ni-content, mechanical distortion, and paragenesis of the metallic particles and iron sulphides, finally the value of H_2O content and the homogenized texture reflect the equilibrated condition. In spite of this wide range of data the petrological classification rise difficulties, because the determination of the homogeneity of olivine and pyroxene based upon a few measurements, and because in the texture characterized by crystalline groundmass and many poorly defined chondrules there are some well-defined chondrules too. *On the basis of all these results the Minnichhof meteorite can be considered as a variety between the L5–L6 types of* VAN SCHMUS–WOOD *two-dimensional classification.*

If we take into consideration the classifying criteria of DODD, VAN SCHMUS and KOFFMAN [1967] we can conclude to a result very similar to the mentioned ones. Summing up the data got from the chemical analyses, the calculated values of Fe°/Fe, SiO_2/MgO, Fe and Fa in olivine, Fe and Fs in pyroxene as well as the data relating to textural features and mineralogical composition the Minnichhof meteorite is an almost equilibrated, metamorphosed chondrite, belonging to the L-group, with an advanced textural integration.

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STUDIES ON PROPERTIES OF MONTMORILLONITE-AMINO ACID COMPLEXES

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ABSTRACT

Three montmorillonite-amino acid complexes (montmorillonite-glicine, montmorillonite-arginine and montmorillonite-lysine) were prepared. The complexes were studied by DTA, X-ray and IR-spectroscopy.

DTA measurements indicate a strong interaction between montmorillonite and organic material. There is a mutual stabilization as evidenced by the shift of peak temperatures of both the structural decomposition of the montmorillonite and the decomposition of the organic material. X-ray measurements support the results obtained by other methods. IR-spectra support the existence of the montmorillonite-organic material interactions. Complex formation brings about such conjugational relationships which result in the appearance of a new absorption band in the IR-spectra.

INTRODUCTION

The well known characteristic of clay minerals is that they readily take up organic matter [EGLINTON, 1969]. Montmorillonite especially features that habit. Montmorillonite stabilizes the absorbed organic material against bacterial degradation, and — under favourable conditions — catalyzes the alteration of the organic substance. However, in the course of the transformation of the organic material, montmorillonite itself turns into a more "illite-like" material [ROLOFF, 1965].

TSUNASHIMA *et al.* [1975] carried out hydrothermal synthesis of montmorillonite-lysine, montmorillonite-glicine and ammonium-mica complexes, respectively. It was proved that the organic component can play an important role in clay formations during the sedimentation and the following diagenetic phases.

This paper reports on the preparation of montmorillonite-amino acid complexes. In the course of the work the amino acid uptake of a given montmorillonite, the type of changes effected by the built-in organic material upon the structure of the montmorillonite, the presence and type of interactions between the clay mineral and the organic matter and their effects upon the thermic properties of the materials involved were studied.

EXPERIMENTAL

Experiments were started with Mád–Koldu (Hungary) bentonite as raw material. X-ray diffractometric and DTA measurements revealed that this material was a mixture of kaolinite and montmorillonite. The raw material was subjected to repeated sedimentation and centrifugation resulting in a practically pure montmorillonite fraction, with grain size of <0.5 micron. Complexes were prepared from that montmorillonite and analytical grade amino acids.

The cation-exchange capacity of montmorillonite was determined by the NH_4Cl-method according to HOFMANN and GIESE. Absorption isotherms were obtained at constant pH, equilibration period, temperature, and amount of the absorbent and only the amino acid concentrations were changed. 2 g absorbents were weighed, mixed with 40 ml distilled water, then agitated for 20 hours on a shaking machine. Different amounts of amino acids corresponding to the different concentration levels were weighed and dissolved in distilled water, then, in turn, added to the montmorillonite suspension. The pH was adjusted and the volume of the suspension was brought to 100 ml. The pH values of the montmorillonite-glycine; montmorillonite-arginine and montmorillonite-lysine complexes were 3,2, 8 and 9 respectively. This procedure was followed by a 24 hours mechanical agitation-equilibration procedure on the shaking machine. The complex was cenrifuged, the supernatant phase separated and sampled for an automatic amino acid analyser to determine the amounts of the unreacted amino acids. The complex obtained was washed four times with 60 ml portions of distilled water. Part of the complex was subjected to DTA, the other part to X-ray investigations. A suspension of 0.5% of the complexes was prepared and the clay mineral-amino acid complex was sedimented onto a sample holder to obtain an oriented sample.

Having determined the conditions of the greatest amino acid uptake on the basis of adsorption isotherms, larger quantities of the complexes were prepared and their DTA curves were recorded. Transformation temperatures of the complexes were determined from the DTA curves, then 2 g samples of the respective complexes were subjected to heat treatment.

Complexes were heated for an hour in N_2 or air stream in tube furnaces at predetermined temperatures. DTA curves of the heat-treated samples were also recorded.

Infrared spectra of both the montmorillonite, the original and heat treated complexes were recorded. 800 mg spectral grade KBr and 3 mg material to be tested were mixed, homogenized and pelletized.

RESULTS

The cation-exchange capacity of montmorillonite was found to be 96 meq/100 g. Based on the adsorption isotherms the maximum amounts of amino acids adsorbed 250 meq/100 g; 560 meq/100 g and 585 meq/100 g in the case of montmorillonite-glicine, montmorillonite-arginine, and montmorillonite-lysine complexes, respectively. However, it should be noted that only apparent absorbed quantities can be spoken about. That means that only a part of these amounts are bound by chemosorption and incorporated into the clay-structure.

The greater part of amino acids is bound only by physical absorption forces to the surfaces. It is evidenced by the fact that having subjected the complexes to vigorous agitation and repeated washings, most of the originally absorbed amino acid is removed. The considerable amount of amino acid thus returned into the solution stabilizes the suspension, so the complex can not be separated during the usual centrifugation time.

X-ray diffraction studies

X-ray diffraction measurements support the results of DTA measurements to be discussed below. The d_{001} values are 12.44 Å; 13.38 Å and 13.48 Å in the case of the montmorillonite-glicine, montmorillonite-arginine and montmorillonite-lysine complexes, respectively. These figures agree well with data in the literature.

DTA measurements

When the complexes obtained by determining the absorption isotherms experiments were studied, it was observed that only a part of the absorbed amino acid is incorporated into the montmorillonite structure. The amount of organic material could be evaluated from the weight losses observed on the TG curves. These calculations may be to a certain extent imprecise, yet this fact does not effect the usefulness of the data obtained. The following cation-exchange capacity values could be derived from the DTA measurements: 33 meq/100 g; 77 meq/100 g and 88.3 meq/100 g in the case of montmorillonite-glicine, montmorillonite-arginine and montmorillonite-lysine complexes, respectively.

DTA measurements made on complexes obtained at different points of the absorption isotherms revealed that due to the 24 hours long shaking process the amount of the absorbed amino acid did more or less approach the exchange capacity value at quite low amino acid concentrations. Based on the DTA curves of the untreated complexes it can be concluded that there is a strong interaction between the montmorillonite and the organic material *(Fig. 1)*. In the case of the montmorillonite-glicine complex the temperature of the second endothermic peak is shifted from 560° C to 590° C, while that of the peak denoting the final structural decomposition is shifted from 870° C to 890° C. Furthermore, an other exothermic peak, so far undetected on the DTA curves of the original montmorillonite, appeared at 920° C.

On the DTA curves of the montmorillonite-arginine and montmorillonite-lysine complexes the endothermic effect corresponding to the decomposition of the montmorillonite structure is almost totally distorted and covered by two intense exothermic peaks appearing between 630° C and 730° C.

The exothermic effect, however, can well be observed at each these complexes at 930° C and 970° C, respectively.

It can be seen from the DTA curves of the heat-treated samples that the peak temperature values corresponding to the structural decomposition of montmorillonite are shifted towards higher temperatures.

The montmorillonite-organic matter interaction manifests itself in increased thermal stability of the organic material. The DTA curves-based thermal stability order is as follows: glicine, lysine and arginine. Thermal cracking of the organic substance bound in the complex occurs at a significantly higher temperature than that derived from the melting point.

IR-measurements

Infrared spectra of the montmorillonite samples agree well with those published by MOENKE [1962], shown in Table 1. It can be seen, that bands belonging to the organic material do not occur equally frequently in the case of different substances *(Fig. 2* and *3)*. Organic bands can best be identified in the case of the montmorillonite-lysine complex, while in the IR-spectra of the other two complexes only one band in each can be identified. The organic bands are as follows:

$\gamma_s > CH_2$ at 1342 cm^{-1}
$\beta_s > CH_2$ at 1405 cm^{-1}
β-NH_3^+ and amide II at 1510 cm^{-1}
ν_s-NH_3^+ at 2950 cm^{-1} and
ν_{as}-NH_2 at 3290 cm^{-1}.

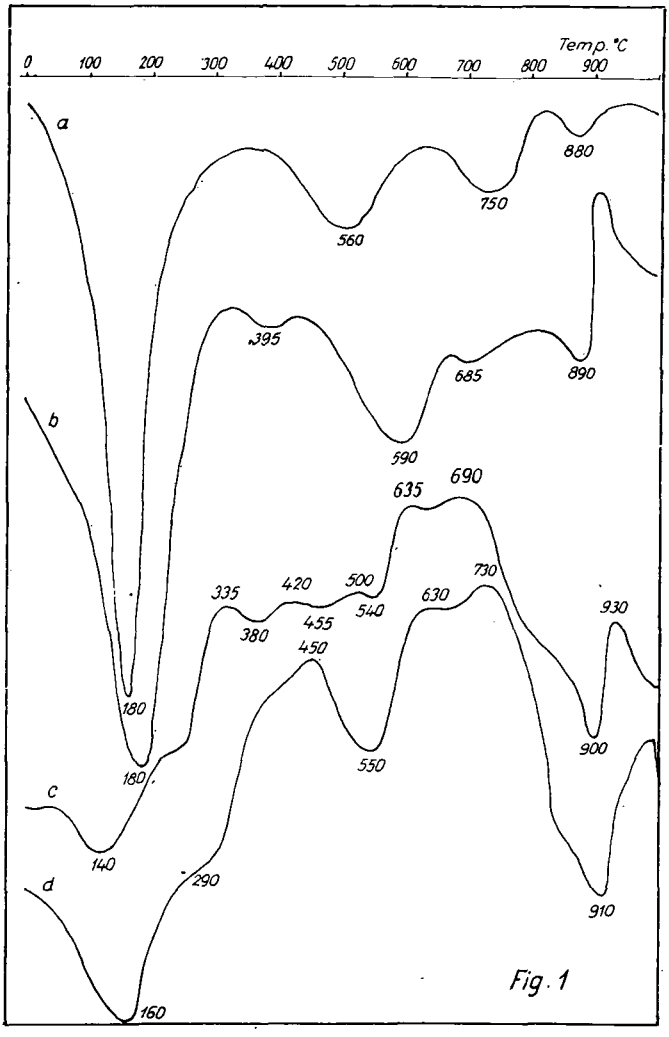

Fig. 1. DTA curves of montmorillonite-amino acid complexes
a) montmorillonite, Mád-Koldu (Hungary)
b) montmorillonite-glycine
c) montmorillonite-arginine
d) montmorillonite-lysine

Bands assigned to the organic substance disappeared in samples subjected to heat treatment at elevated temperatures. Treating the samples at 600° C and 800° C for 1 hour a number of bands corresponding to montmorillonite fundamental vibrations disappeared (at 430 cm^{-1}, 353 cm^{-1}; 925 cm^{-1}, and 3635 cm^{-1}), while new bands appeared (at 740 cm^{-1}, and 810 cm^{-1}).

There is a dublette at 2370 and 2350 cm^{-1} in the IR spectra of each complex; a peak nonexistent in the IR spectrum of the original montmorillonite but always present in the complexes. It is probably due to the special conjugation conditions brought about by the complex formation.

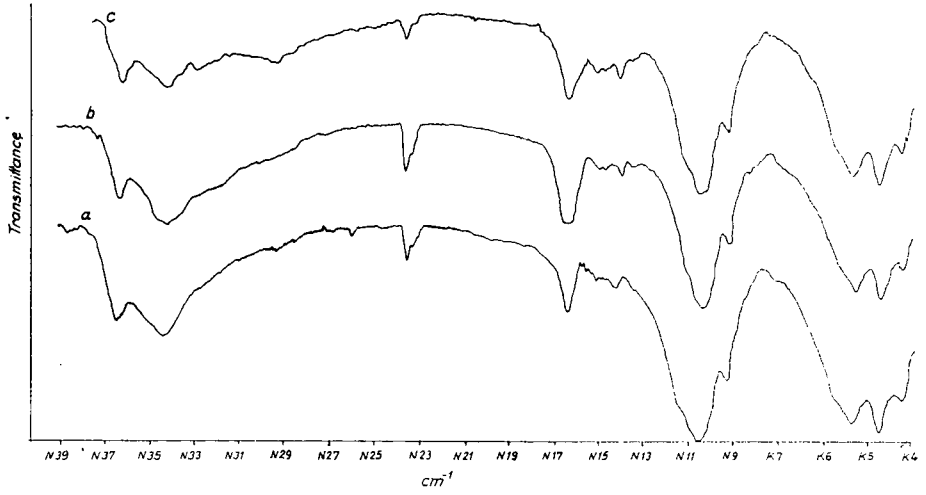

Fig. 2. Infrared spectra of montmorillonite-amino acid complexes
a) montmorillonite-glycine
b) montmorillonite-arginine
c) montmorillonite-lysine

TABLE 1

Major bands in the IR-spectra of montmorillonites

Group; type of vibration	Wave number range cm^{-1}	Montmorillonite bands in MOENKE's samples cm^{-1}	Montmorillonite bands in MÁD-KOLDU samples cm^{-1}
Fundamental vibrations of the SiO_4 tetrahedron and AlO_6 octahedron	(600—350)	470 525 630	426 475 530
ν_s (Si—O)—Si	850—800	850	810
ν_{as} (Si—O)—$Al^{(6)}$	1000—830	920	920
ν_{as} (Si—O)—$Al^{(4)}$	1055—980	1040	1040
ν_{as} (Si—O)—Si	1120—1070	1100	1100
β—OH	1680—1600	1650	1650
ν H—O—H	3600—3000	3430	3470
ν —OH	3760—3520	3628	3630

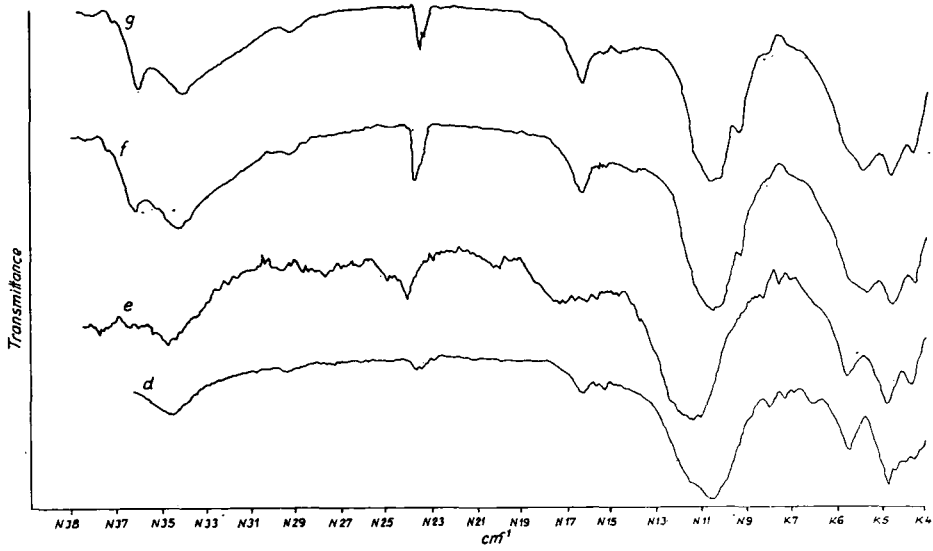

Fig. 3. Infrared spectra of heat treated samples
d) montmorillonite-lysine (800 °C)
e) montmorillonite-lysine (600 °C)
f) montmorillonite-arginine (420 °C)
g) montmorillonite-arginine (320 °C)

SUMMARY

Results of DTA and IR measurements yield valuable information relating to the montmorillonite-amino acid complexes. There is a mutual stabilization between the organic substance and montmorillonite resulting in 20–40° C higher temperature values for the structural changes of montmorillonite and increased thermal stability of the amino acids. Since the remaining products and none of the gases evolved could be studied by the DTA investigations, it would be proper to extend these tests and to study the gases evolved as well.

Bands corresponding to the organic material and montmorillonite occur at separate wave numbers. There are vibrations in the 2370–2350 cm^{-1} range, which can be assigned to the complex formation.

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CONTRIBUTIONS TO THE KNOWLEDGE OF THE HUNGARIAN OIL SHALE KEROGEN I

PRELIMINARY REPORT ON THE RESULTS OF THE PYROLYSIS AND SELECTIVE OXIDATION

M. HETÉNYI, K. MAITZ and É. TÓTH

INTRODUCTION

"As petroleum is a natural resource that is being rapidly consumed, there are widespread efforts to seek possible substitutes for it. The reserves of oil shale stand out as a very important source of substitutes for petroleum" [YEN and CHILINGARIAN, 1976]. Recently, the energy crisis and the increasing oil prices throw gradually light upon the problem, i.e. to find other energy resources. Thus, in addition to the productive oil shale occurrences, the research of smaller occurrences of local importance may arise partly from economic, partly from prospecting points of view. It can be assumed that the results of scientific researches explore new profitable possibilities of application which are recently more or less unknown. The investigations of FOMINA [1968] show this trend, resulting the chemical-industrial and agricultural utilization of oxidation products of the Estonian kukersit kerogen.

GENERAL REMARKS ON THE OCCURRENCE

In Hungary oil shale was found in a Pliocene basin in 1973 by the experts of the Hungarian State Geological Institute [JÁMBOR *et al.* 1975]. The rock filling up the basin is greyish-green, apparently light and of lamellar stratification, it is of characteristic smell and combustible. 10 to 70 per cent of it is of algal origin. Its inorganic components are clay minerals, fine-grained sand deriving from the region of the neighbouring basalts, and limy material. Out of its organic components the *Botryococcus braunii* planktonic alga is predominant, further the pollens of terrestrial plants are frequent [JÁMBOR *et al.* 1975]. According to the detailed investigations of algal and pollen remnants these oil shale strata were formed in the Pliocene, in semihaline water of 10 to 12° C of a crater lake of appropriate pH, salinity and temperature, and which proved to be suitable to abundant production of protophytes. Since the protophytes in question are highly sensible to the factors mentioned above, their large-scale destruction was probably caused by cold-water inundations and by the change of salinity. On the basis of the calcite/aragonite ratio determined by means of X-ray diffractometer two sedimentation cycles can be distinguished in the basin: the boundary of the cycles is in a depth of 19 to 20 metres, the start of the first cycle is probable in a depth of about 40 metres [MEZŐSI, 1976].

Part of the oil shales and of other sediments, i.e. the bitumen fraction is soluble in the common organic solvents. The other part is insoluble in organic solvents, this is usually called kerogen. The bitumen fraction of oil shales is max. 20 per cent of the total organic matter content, however, its value is less, in general. Consequently, the major part of the organic matter is kerogen, thus if the oil shale is to be

characterized, its kerogen should be studied. Unfortunately, while the bitumen fraction can be easily separated by extraction, the isolation of kerogen is troublesome and hardly can be carried out without the decomposition of the original matter by using chemical methods. This is why efforts are made to characterize the kerogen content of the oil shales, and which do not need the preliminary removal of inorganic components, e.g. measurement of reflectance of vitrinite, determination of reduction capacity of the organic matter by means of selective oxidation, pyrolysis, etc.

The results of the preliminary investigations on the kerogen of the Hungarian oil shale are summarized. These investigations were carried out according to the scheme shown in *Fig. 1.* It is to be noted here, that certain measurements were carried out only in some representative samples. In Table 1 some characteristics of the samples investigated are demonstrated.

The peculiarities of the oil shale kerogen depend on the diagenesis, on the chemical features of the sedimentary environment, on the geological conditions (closed bay, or inland sea, etc.) and on the biological source matter. The data of the elemental analysis are used to characterize these materials (H, C, N, O, S), or as an approach, the hydrogen and carbon content, resp., the H/C atomic ratio is given.

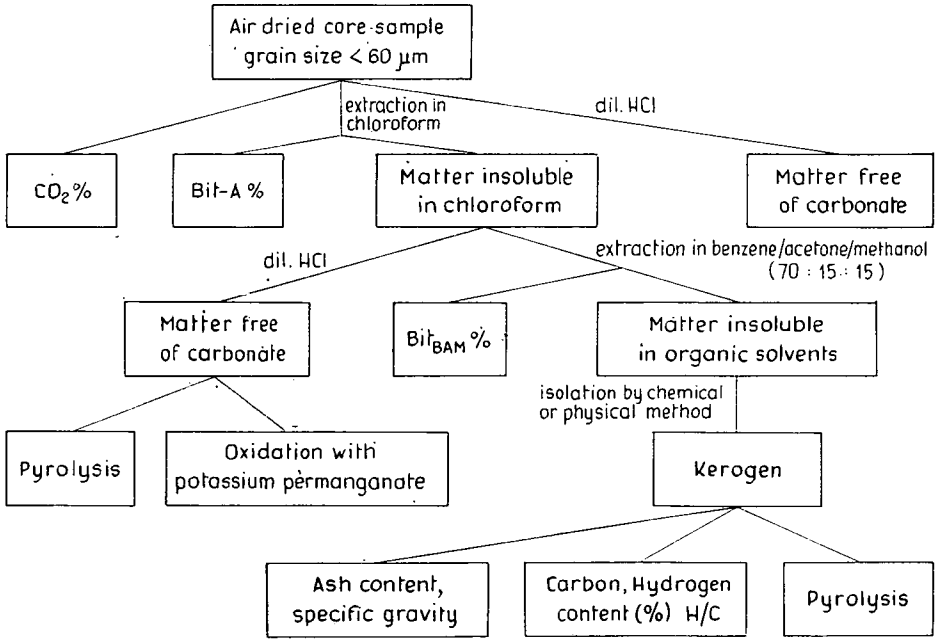

Fig. 1. Investigation scheme of organic matter of the Hungarian oil shale.

Having removed the bitumen fraction from the samples by means of repeated extraction, the kerogen has been isolated. This has been carried out by physical methods in most of the cases: flotation in calcium chloride solution in order to avoid the chemical transformation of the organic matter. The concentrate, however, will contain inorganic matter of small quantity, especially in the samples of low organic carbon content. The samples of C_{org} content less than 10 per cent do not produce kerogen concentrate of required purity, their data are published only

TABLE 1

Carbon dioxide (CO_2), organic carbon content (C_{org}), organic matter soluble in chloroform (Bit—A) and organic matter soluble in organic solvents (Bit_{BAM}) of samples from the borehole Put-7

Sample No	Depth m	CO_2 (%)	C_{org} (%)	Bit—A (%)	Bit_{BAM} (%)
5	4,8— 5,4	18,5	2,27	0,14	0,07
7	6,0— 6,5	17,3	1,69	0,11	0,08
11	8,0— 8,5	11,2	6,88	0,61	0,19
14	9,5—10,0	9,5	11,30	1,40	0,54
18	11,5—12,0	16,1	7,69	0,84	0,55
22	13,4—14,0	18,0	8,94	1,80	0,43
24	14,5—15,0	6,7	32,56	3,61	2,04
26	15,5—16,0	3,7	45,61	5,12	1,73
27	16,0—16,5	4,3	45,68	3,95	2,01
28	16,5—17,0	4,8	28,89	4,98	1,54
32	18,5—19,0	15,5	11,44	1,35	0,79
33	19,0—19,5	21,4	9,89	0,55	0,49
35	20,0—20,5	22,1	11,80	1,15	0,32
38	21,5—22,0	14,7	16,74	3,99	1,18
42	23,5—24,0	17,4	18,64	1,56	0,95
45	25,0—25,5	11,8	30,09	2,99	0,85
49	27,0—27,5	16,1	21,16	4,77	1,49
55	30,0—30,5	22,2	8,74	0,47	0,05
59	32,0—32,5	21,5	10,54	0,67	0,27
62	33,5—34,0	23,7	6,80	0,53	0,38
69	37,0—37,5	24,6	5,00	0,28	0,17
73	39,0—39,3	29,8	6,15	0,43	0,23

for comparison. In the samples No. 14 and 18, the carbon content of which proved to be relatively low, kerogen was isolated by chemical method.

The carbon and hydrogen contents as well as the H/C ratios of the concentrates are shown in Table 2. The average value of the *H/C* ratio is 1.77. It is low in case

TABLE 2

Carbon, hydrogen content, H/C atomic ratio and method of isolation of the kerogens

Sample No	Method of the isolation	C (%)	H (%)	H/C atomic ratio
14	chemical	67,5	9,5	1,6
18	chemical	73,0	8,5	1,4
24	physical	72,9	11,6	1,9
26	physical	69,2	11,2	1,9
27	physical	73,6	11,7	1,9
28	physical	76,7	12,7	2,0
32	physical	72,6	11,7	1,9
33	physical	74,5	11,5	1,8
35	physical	80,5	12,5	1,8
38	physical	72,9	10,0	1,7
42	physical	69,6	10,2	1,8
45	physical	73,5	10,5	1,7
49	physical	65,6	9,8	1,8
55	physical	17,6	2,8	1,9
62	physical	57,3	8,1	1,7
69	physical	12,4	1,8	1,7

of the two chemically isolated samples (1.4–1.6), in case of the other no significant deviation is found.

The *H/C* ratios of the Hungarian oil shale were compared with those of other kerogens, the source matter of which has been also *Botryococcus* alga. In Table 3 the values from the literature as well as that of our samples are shown. The H/C ratio of the Hungarian oil shale agrees with that of the Coorongite of Australia. Coorongite represents a rather early stage in the formation of the algal kerogen, it is the occurrence of *Botryococcus braunii* in form of free colonies. "Coorongite should be considered similarly to the peat state when the algal shales are coalified" — writes CANE [1969].

TABLE 3

Age, precursors, carbon, hydrogen content and H/C atomic ratio of some kerogens

Name and location of kerogen	Age	Precusor	C (%)	H (%)	H/C
Torbanite Scotland	Permian	*Pilaminima P. scotica (Botryococcus)*	79,1	9,8	1,49
Marahunite Brazil	Tertiary	*Botryococcus braunii*	58,9	5,5	1,12
Autun Boghead France	Permian	*Pila bibractensis (Botryococcus)*	76,9	10,4	1,62
Alginite Australia	Permian	*Pila reinschia (Botryococcus)*	72,5	9,1	1,28
Coorongite Australia	Recent	*Elasophyton coorongina (Botryococcus)*	76,1	11,2	1,77
Alginite Hungary	Pliocene	*Botryococcus braunii*			1,77

INVESTIGATION OF ORGANIC MATTER BY PYROLYSIS

To study the character, composition and degree of diagenesis of the organic matter, pyrolysis is one of the most wide-spread method. Similarly to other methods, this method is also unable to describe alone the structure and evolution of organic matter of the oil shales but provides satisfactory informations as a first approach.

Pyrolysis is an artificial evolution process, i.e. the evolution taking place in the nature at low temperature, high pressure and during long time can be modelled by pyrolysis carried out at higher temperatures in laboratory conditions. TISSOT *et al.* [1974] evidenced that the high temperature eliminates the time and pressure factors of evolution. The conditions of heating (vacuum or inert atmosphere, solvent medium, temperature) determine the mechanism of the process, the quality and quantitative distribution of its products. Certain part of the pyrolysis investigations try to explore the structure of kerogen. The different groups and radicles produced by the low temperature pyrolysis are detected by gas chromatographs or mass spectrometers. When applying low temperature (200 to 300° C) the paraffin or aromatic

character of the organic matter can be determined, further data can be obtained concerning the gas and oil potential, and the kinetics of the decomposition process can also be studied [CANE, 1948; DI RICCO and BARRICK, 1956; GIRAUD *et al.* 1970].

By means of higher temperature pyrolysis (500 to 600° C) the evolution processes can be followed [TISSOT *et al.*, 1974; GIRAUD *et al.*, 1970].

GRANSCH and EISMA [1966] characterized the source and evolution degree of the kerogen by the C_R/C_T quotient were C_R denotes the carbon content after the pyrolysis at 900° C and during 90 minutes and C_T is the total carbon content. GIRAUD *et al.* [1970] measured the C_R/C_T quotient at 500°C and this was called *degradation factor* (T_D). These methods are favourable since the preliminary removal of the inorganic compounds can be neglected.

Our investigations were carried out according to GRANSCH and EISMA [1966] with the only modification that pyrolysis was performed not only at 900° C but also at 300, 400, 500 and 600°C. Pyrolysis was carried out in an electric furnace, in a quartz tube. The furnace was heated to the required temperature, then the weighted sample was taken into the furnace where it was pyrolized during 90 minutes in nitrogen atmosphere. Cooling to room temperature was carried out also in nitrogen atmosphere, then the sample was weighted again. The carbon contents obtained before and after pyrolysis at different temperatures, respectively, were determined (signs of the values: C_T, C_{300}, C_{400}, ... C_{900}). The degradation factor (T_D) was computed for all temperatures: $T_{300}=C_{300}/C_T$, etc. (Table 4). According to GRANSCH and EISMA [1966] in case of alginite the value of C_{900}/C_T measured at 900° C is 0.05–0.27, in case of peat, lignite and humic coals 0.60–1.00. In the samples investigated containing alginite the C_{900}/C_T value proved to be 0.09–0.24, in samples

TABLE 4

Degradation factors $\left(T_D = \frac{C_R}{C_T}\right)$ *measured by pyrolysis at different temperatures*

Sample No	T_{300}	T_{400}	T_{500}	T_{600}	T_{900}
5	0,83	0,83	0,77	0,54	0,34
7	0,86	0,86	0,74	0,49	0,42
11	0,96	0,95	0,86	0,32	0,16
14	0,98	0,94	0,81	0,28	0,24
18	0,99	0,94	0,79	0,49	0,35
22	0,92	0,87	0,81	0,40	0,32
24	0,98	0,83	0,71	0,15	0,11
26	0,95	0,95	0,81	0,17	0,12
27	0,96	0,96	0,86	0,13	0,09
28	0,96	0,96	0,96	0,17	0,15
32	0,96	0,91	0,80	0,31	0,10
33	1,00	0,98	0,82	0,51	0,27
35	1,00	0,89	0,84	0,49	0,23
38	0,95	0,91	0,81	0,23	0,14
42	0,97	0,95	0,82	0,33	0,20
45	0,96	0,95	0,87	0,18	0,13
49	0,97	0,98	0,74	0,21	0,16
55	0,92	0,90	0,78	0,53	0,28
59	0,93	0,91	0,75	0,45	0,40
62	0,95	0,90	0,74	0,48	0,29
69	0,99	0,99	0,71	0,48	0,14
73	0,92	0,92	0,85	0,59	0,41

rich in organic matter around 0.10. The T_D values were plotted as a function of temperature and some characteristic curves are seen in *Fig.* 2. It is characteristic of all the samples that the transformation of the organic matter follows mostly between 500 and 600° C but in case of a few samples this section is steeper than in case of others. Further differences are found, *i.e.* in certain samples pyrolysis seems to be completed at 600° C, in others the curve is steep also between 600 and 900° C. To make easier the comparison, the characteristic curves of the samples were described by a figure, thus the rise of the curves between 500 and 600° as well as between 600 and 900° C being apparently characteristic of the samples were also determined

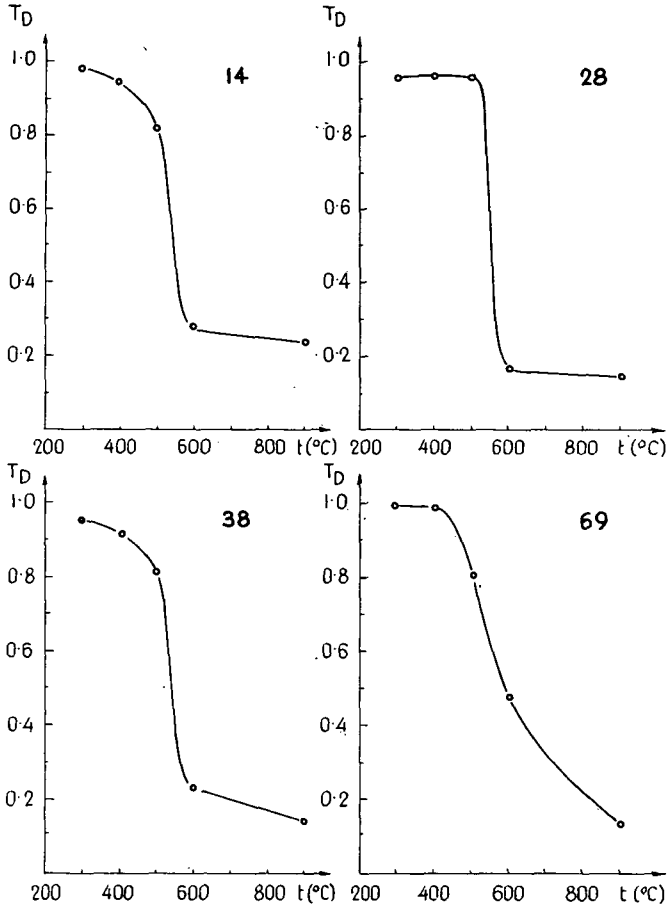

Fig. 2. Pyrolysis curves of some samples of the borehole Put-7. $T_D = C_R/C_T$ degradation factor t (°C) temperature of pyrolysis.

(marked by m_1 and m_2), finally their quotient (m_1/m_2) was formed. On the basis of these values the investigated samples can be assigned to three groups (Table 5). In the first group the value of m_1/m_2 is 2.0 to 7.0; in the second group it is 10.4–19.3 while in the third group it is greater than 33. The distribution of the samples among the three groups seems to be concordant with the geological view developed on the

TABLE 5

Slope of the pyrolysis curves of the samples from boreholes Put-7

I		II		III	
Sample No.	$\frac{m_1}{m_2}$	Sample No.	$\frac{m_1}{m_2}$	Sample No.	$\frac{m_1}{m_2}$
69	2,0	11	10,4	49	33,1
55	3,0	7	10,9	26	40,0
5	3,5	42	12,0	14	40,8
33	3,9	22	15,4	24	43,0
35	4,1	59	18,8	45	43,1
73	4,3	38	19,3	27	56,2
18	6,0			28	>60,0
32	7,0				

basis of the stratigraphic and X-ray diffractometric investigations. As it has been emphasized in the introduction, the Hungarian oil shale was formed in a small bay surrounded by the ring-shaped tuff barriers of a volcanic crater [JÁMBOR *et al.*, 1975]. Smaller cold-water inundations caused periodic temperature and salinity changes in several phases and this had been responsible for the destruction of the *Botryococcus* colonies [MEZŐSI, 1976]. Out of our samples those of No. 24, 26, 27, 28 and 14 derive from the depth corresponding to such inundation periods. This may explain the fact that the sample No. 14 of relatively low carbon content (C=11.3%) and the samples of high carbon content (C=20.45%) form the third group characterized by the highest m_1/m_2 value. Samples being assigned to the first group ($m_1/m_2<7$) derive from the boundaries of the sedimentation cycles, their organic carbon content is low and is composed more or less of coalified plant remnants.

The transitional samples lying between the two extreme values belong to the second group.

Comparing our measurement results with the data of the literature, e.g. with those of TISSOT *et al.* [1974] it is apparent that according to the authors above significant change follows during pyrolysis between 400 and 500° C while according to our data this occurs only between 500 and 600° C. It was thought that the difference can be attributed to the inorganic components. To prove this assumption the pyrolysis was carried out on some kerogen concentrates by means of the method described above. The specific gravity and ash content characteristic of the purity of the concentrates as well as the degradation factors determined by pyrolysis are shown in Table 6. Since the kerogen investigated was concentrated by means of physical

TABLE 6

Specific gravity, ash content and degradation factors of the kerogens isolated from samples of the borehole Put-7

Sample No.	T_{300}	T_{400}	T_{500}	T_{600}	T_{900}	Specific gravity	Ash content %
26	0,91	0,38	0,07	0,06	0,03	0,89	10,56
28	0,91	0,40	0,05	0,05	0,03	0,99	7,67
38	0,92	0,47	0,05	0,04	<0,03	1,12	9,18
55	0,94	0,55	0,47	0,36	0,09	1,65	64,7
69	0,91	0,78	0,70	0,44	0,24	1,48	71,10

method, in case of the samples No. 55 and 69 of lower organic matter content the concentration is only partial, the specific gravity and ash content are fairly high. In case of these samples the rise of the curves is similar to that of the original matter, the main thermal process takes place also between 500 and 600° C and 600 and 900° C, resp. In case of the samples No. 26, 28 and 38 the concentrate is somewhat purer, and pyrolysis seems to be completed at 500° C and this is in accordance with the data of references. On the basis of this measurement series it is believed that the difference of about 100° C is caused by the inorganic components and this problem will be investigated in detail.

CLASSIFICATION OF THE ORGANIC MATTER BY MEANS OF SELECTIVE OXIDATION METHOD

On the basis of the wide-spread methods applied in carbon chemistry numerous authors studied the effect of different oxidizing media on oil shales, especially that of potassium permanganate. "This research had the very important result that it clearly differentiated oil shale kerogens into two types depending upon whether benzenoid acids were formed or not. Further research has led to the belief that at least two types of kerogen are likely to be present in oil shales: 1. an algal portion which may or may not be oxidation resistant and 2. a quasi-aromatic portion the nature of which is more humic or coaly" [CANE, 1976]. As an improvement of the method and to avoid the further oxidation of the transitional products either special oxidation medium is used, e.g. the acetonic solution of potassium permanganate of STEFANOVIČ and VITOROVIČ [1959] being specific of double bonds, or the oxidation is carried out step by step. To classify the petroleum source rocks and to estimate the oil potential an empirical formula was elaborated by KHALIFEH and LOUIS [1961]; the method is based on the analysis of the non-oxidized residue of the step by step oxidation. Acidic potassium permanganate was used as oxidizing medium, and after oxidation the reduction capacity (PR) and carbon content (C) of the organic matter remained in solid phase were determined. In case of our investigations this latter method has been used. Samples were prepared in the manner shown in *Fig. 1.* Since the matter of about 3 per cent organic carbon content is most favourable for the measurements, the oil shale was "diluted" by kaolinite by five to ten times. (The selective oxidation was carried out, of course, also by kaolinite and while no change in the parameters needed by our measurements could be measured during the process, it proved to be a suitable diluting medium.) The quotient of reduction capacity and organic carbon content ($C_R = PR/C$) was plotted against the organic carbon content. Seven samples were chosen, three from the second sedimentation cycle, the samples No. 24 and 28 being characterized by numerous large-sized *Botryococcus* colonies, and finally the sample No. 32 deriving from the lower boundary of the sedimentation cycle. The carbon content of this latter sample is lower and contains a few coalified plant fragments. In the first sedimentation cycle samples No. 38, 42 and 49 represent the relatively enriched organic matter content, out of the investigated samples that of No. 69 is of lowest carbon content ($C = 5\%$).

The $C_R - C$ curves obtained as a result of selective oxidation are of the same rise in the sample No. 24 and 28 out of the seven investigated samples *(Fig. 3a)*, and they are nearly parallel with the abscissa. The curve of the sample No. 38 seems to be divided into two sections, i.e. up to about $C = 50\%$ it is parallel with the abscissa, then it turns steeply downward *(Fig. 3b)*. The curves of the samples No. 42 and 49

(Fig. 3c) incline gradually downward but after about C=60% the sudden decrease of the C_R-values follows. The change of the C_R values of the samples No. 32 and 69 is nearly linear as a function of C. The reduction capacity of the samples decreases in the order described above, *i.e.* initially the *Botryococcus braunii* remnants determine the character of the organic matter, and parallel with their gradual decrease the coalified plant remnants become predominant out of the components of the organic matter. Consequently, the rise of the curves will be determined by the maturity of the organic matter and not by the absolute value of the organic carbon content, since regarding *e.g.* the sample No. 32 (C=11.44%) lies closer the sample No. 38 (C=16.74%) than the sample No. 69 (C=5.00%). Nevertheless, the reduction capacity of the samples No. 32 and 69 shows similar features as a function of the carbon content while the sample No. 38 is transitional between the sample No. 24 and 28 rich in organic matter and those of No. 42 and 49.

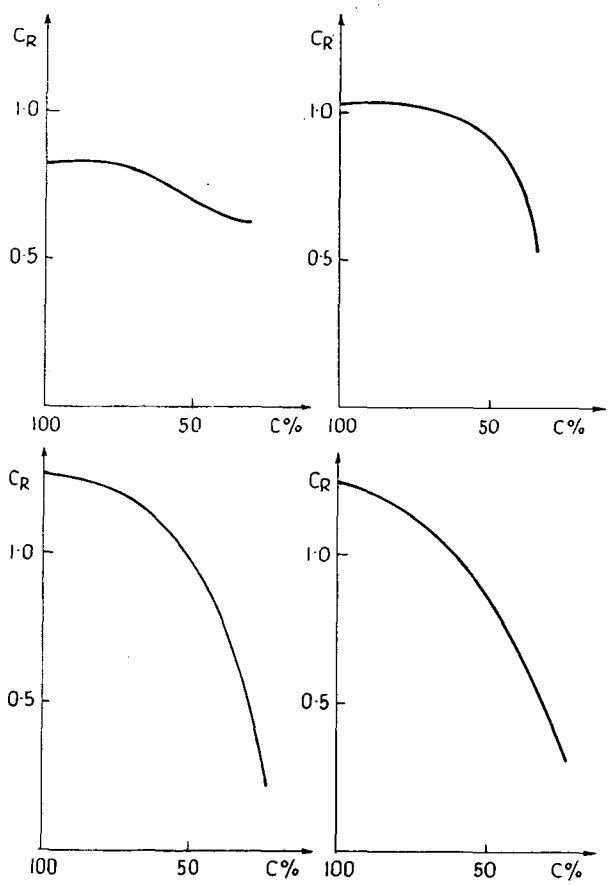

Fig. 3. Change of reduction capacity of the organic matter during selective oxidation in some samples of the borehole Put-7.
a) samples No 24 and 28;
b) sample No 38;
c) samples No 42 and 49;
d) samples No 32 and 69.

SUMMARY

The kerogen of the Hungarian oil shale tried to be characterized by several methods.

Kerogen was isolated, its carbon and hydrogen contents were measured, its H/C ratio was calculated and these were compared with the same data of other oil shales known from literature. The H/C ratio depends on the maturity of the kerogen and on its diagenetic state. The source material as well as the sedimentation conditions and chemical parameters of the environment also affect on this ratio. According to GRANSCH and EISMA [1966] in case of alginites the H/C ratio moves between 1.4 and 1.8. In case of the Hungarian oil shale this ratio proved to be 1.77, in average. Lower values were obtained in case of chemically isolated samples (1.4–1.6) and somewhat higher in case of samples isolated by means of physical methods. When comparing these samples with other alginites the source matter of which was also Botryococcus alga, the obtained average value is close to that of the Australian coorongite.

Further, methods were applied to determine the character of the organic matter which do not need the preliminary separation of the organic matter, but after removing the bitumen fraction and the carbonates, the measurement can be carried out just on the original rock. Though no information was obtained regarding the chemical composition and structure of kerogen by means of these methods, so these provide only preliminary informations, distinction can be made within one borehole in the character of the organic matter (in our case also within 40 metres).

Pyrolysis is such a method which is suitable to describe the source material and degree of evolution of the kerogen. The samples were pyrolised step by step at 300, 400, 500, 600 and 900° C. At 900° C the ratio of the bound carbon content to the total carbon content is 0.05 to 0.27 in case of alginites [GRANSCH and EISMA, 1966]. In the investigated alginite-bearing samples the C_{900}/C_T value proved to be 0.09 to 0.24, in the samples most abundant in organic matter around 0.10. The degradation factors determined at different temperatures were plotted against the temperature and on the basis of the rise of the curves characterized by the slope of each curve, the samples could be divided into three groups. The samples containing only coalified plant remnants and the samples of algal origin and rich in organic matter belong to the two extreme groups. The second groups contains the samples of transitional character. The samples assigned to the first group correspond to the depth intervals determining the start and end of each sedimentation cycle. The samples of the third group derive from a depth where, as a result of the gradual changes of chemical parameters of the crater lake, the Botryococcus colonies were destroyed in large masses.

The character of the organic matter can be described by selective oxidation as a first approach, since the resistance capacity of the organic matter against the oxidizing agents is in relationship with the kerogen structure and source material and the diagenetic process affects them only to a smaller extent [YEN and CHILINGARIAN, 1976]. On the basis of the C_R—C curves the samples containing different kinds of organic matter can be decisively separated and the curves of the samples of the two sedimentation cycles rich in organic matter (samples No. 24, 28, resp. 42, 49) are also different. On the basis of the sedimentological investigations the following difference can be found among the samples mentioned above: in the depth interval of 13 to 19 m (samples No. 24 and 28) the Botryococcus colonies occur in large masses, while the samples No. 42 and 49 are characterized by lamellar formation and only Botryococcus colonies of smaller size can be observed in them. Further

investigations extending also over the analyses of the oxidation products will throw light upon the problem that the differences in the features of the samples can be attributed to the smaller differences in the kerogen structure, or this is caused by the "advantage" of the investigation method (which may occur here as a failure) that the inorganic components were not preliminarily separated from the organic matter.

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CONTRIBUTIONS TO THE KNOWLEDGE OF THE HUNGARIAN OIL SHALE KEROGEN II

RESULTS OF PRELIMINARY DTA AND IR-INVESTIGATIONS ON THE KEROGEN OF THE OIL SHALE OCCURRENCE AT PULA

GY. GRASSELLY, M. BERTALAN and CS. SAJGÓ

INTRODUCTION

The geological conditions and general characteristics of oil shales discovered in 1973 in Transdanubia (Hungary) were first reported by Á. JÁMBOR and G. SOLTI [1974]. The petrographical description of the oil shales was carried out by CS. RAVASZ [1974]; J. MEZŐSI and M. MUCSI [1976] dealt with the sedimentology and mineralogy of the occurrence at Pula (Put-7 boring). According to the palynological investigations [Hungarian Geological Institute, E. NAGY, 1974] most part of the insoluble organic material of the sediments consists of *Botryococcus braunii* changing in amount layer by layer besides other organic material (pollens, coalified plant remnants etc.).

In connection with the detailed investigation of the oil shale occurrence of Upper Pannonian age at Pula the necessity of the examination of the kerogen content has also arisen. The following questions may arise: *a)* to what extent the peaks in the DTG and DTA curves attributable to the organic material are modified by inorganic components in the derivatograms of average samples; *b)* what kind of differences appear in the derivatograms of kerogen concentrates isolated by using physical and chemical methods, respectively, *c)* whether or not the type of kerogen from the different layers of Put-7 boring shows changes depending on the depth, sedimentation cycle, rock type; *d)* in what manner the single steps of thermal decomposition of the kerogen concentrates can be characterized by IR-investigations in comparison with the results of derivatographic examinations.

CHARACTERIZATION OF SAMPLES INVESTIGATED

The samples used for investigations from the borehole Put-7 in neighbourhood of village Pula are given in Table 1.

The air dried samples were ground and the carbonate CO_2 content of the samples was determined by gasvolumetric method. The total organic carbon content was determined by ignition in oxygen stream in portion of samples free of carbonate. The soluble organic material of the samples was extracted in a SOXHLET-apparatus in two steps, first by chloroform applying an exhaustive extraction, later continuing it by extraction with benzene-acetone-methanol mixture. Data obtained are shown in Table 2.

Fig. 1 shows the change of total carbon content through the section of boring Put-7, and that of the value of the FISCHER-assay, the latter expressed in per cent. It may be established that a fairly strong positive correlation exists between the two series.

TABLE 1

Samples investigated

Sample	Depth	Characterization of the layer
A—14	9,5—10,0 m	alginitic aleurite
A—15	10,0—10,5	aleuritic alginite
A—18	11,5—12,0	thin-layered type
A—24	14,5—15,0	rich in alginite
A—26	15,5—16,0	
A—27	16,0—16,5	without fine-layering, with a few carbonate
A—28	16,5—17,0	
A—32	18,5—19,0	alginite in a lesser amount, without layering, with more carbonate
A—33	19 0—19 5	
A—35	20 0—20 5	alginite with alternating carbonate content, built up from layered sections and sections without layering
A—38	21 5—22 0	
A—42	23 5—24 0	
A—45	25 0—25 5	rich in alginite, without fine-layering
A—49	27.0—27,5	alginite with carbonate, layered type
A—52	28,5—29,0	alginite with more carbonate, layered type
A—73	39,0—39,3	alginite with more carbonate without layering

TABLE 2

Some characteristic data of samples

Sample	Carbonate CO_2 %	ΣC_{org} (%)	Organic material soluble (%) in chloroform		in solvent mixture		total soluble
A—14	9,5	11,30	1,40	+	0,54	=	1,94
A—15	8,2	11,70	2,34	+	0,43	=	2,77
A—18	16,1	7,70	0,84	+	0,55	=	1,39
A—24	6,7	32,56	3,61	+	2,04	=	5,65
A—26	3,6	45,61	5,12	+	1,73	=	6,85
A—27	4,3	45,68	3,95	+	2,01	=	5,96
A—28	4,8	28,89	4,99	+	1,54	=	6,53
A—32	15,5	11,44	1,35	+	0,79	=	2,14
A—33	21,4	9,89	0,55	+	0,49	=	1,04
A—35	22,1	11,80	1,15	+	0,32	=	1,47
A—38	14,7	16,74	3,99	+	1,10	=	5,09
A—42	17,4	18,64	1,56	+	0,95	=	2,51
A—45	11,8	30,09	2,99	+	0,85	=	3,84
A—49	16,0	21,16	4,77	+	1,49	=	6,26
A—52	17,0	8,52	1,06	+	0,48	=	1,54
A—73	29,8	6,15	0,43	+	0,23	=	0,66

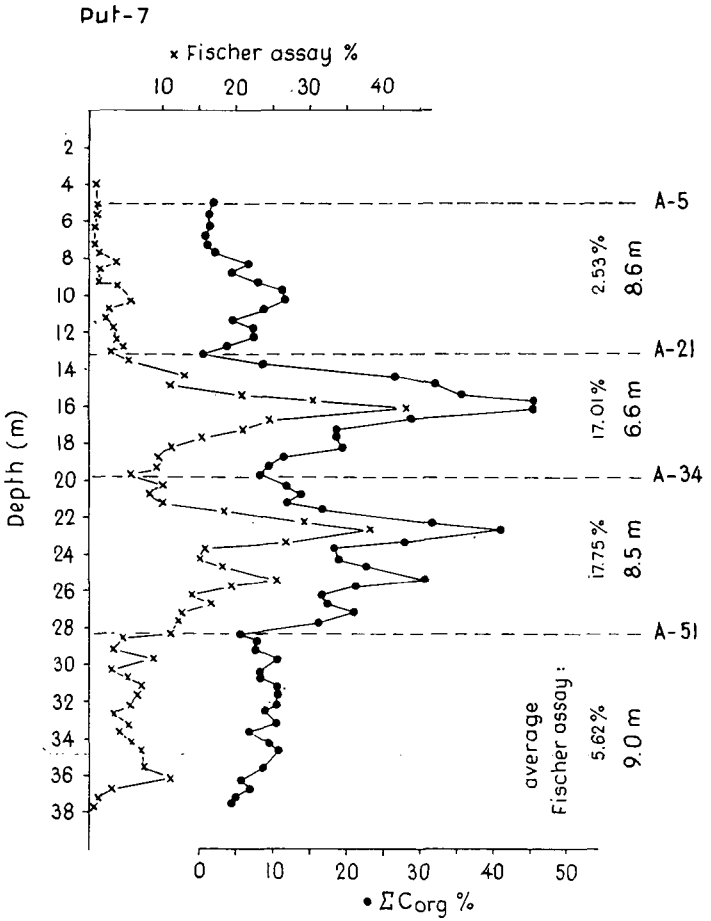

Fig. 1. Change of the ΣC_{org} content and the FISCHER-assay in the section of boring Put-7.

DERIVATOGRAPHIC INVESTIGATIONS

Derivatographic investigation of original samples

Investigations were carried out by an ERDEY–PAULIK–PAULIK Derivatograph in air recording the T, DTA, DTG and TG curves simultaneously under the same conditions. Heating rate 10° C/min.

Peak temperatures in DTG and DTA curves of the derivatograms of the original samples are comprised in Table 3 and 4, respectively.

The temperature of peak I (loss of water) changes from 100° C to 130° C, peak II (between 180–280° C), peak III (between 280°–360° C) and peak IV (450°–480° C) represent the steps of the combustion of the organic material of the sample whereas further maxima denote partly the decomposition of phyllosilicates and that of carbonates. At any rate, in the interval of peaks V and VI an overlapping of decomposition processes of organic material and clay minerals may exist.

TABLE 3

Temperature of maxima in the DTG curve of the original samples

Sample	Temperature of DTG maxima							
	I	II	III	IV	V	VI	VII	VIII
A—14	130	280		460	520		700	820
A—15	125	265		460	520		680	800
A—18	130	280	(360)	(480)			700	840
A—24	100	220	300	465		(580)	(720)	820
A—26	100	220	290	460			(720)	810
A—27	100	180	280	460			760	
A—28	100	200	290	465				810
A—32	100	280		460	530		720	850
A—33	120	260		460	520	560	735	870
A—35	110	260		460	520	570	720	830
A—38	100	230	300	465		570	730	820
A—42	100	240	305	460	(540)		800	840
A—45	110	210	280	460		560	720	820
A—49	100	260	(310)	465	530		720	840
A—52	120	270		(460)			720	840
A—73	120	280		(450)			750	870
Av.:	110°	245°	301°	461°	526°	568°	726°	832°

Values in parenthesis indicate very weak effects.

TABLE 4

Peak temperatures in the DTA curve of the original samples

Sample	Temperature of DTA peaks						
	I	II	III	IV	V	VI	VII
A—14	155	330	(400)		495	(520)	
A—15	140	320	(420)		500		
A—18	150	335	(400)		500		560
A—24	105	270			490	535	(550)
A—26	115	275	435		480	520	
A—27	120	260			480		580
A—28	105	260	400	440	480	520	(550)
A—32	140	320			485	(510)	
A—33	145	320	(395)		485	(515)	(560)
A—35	130	300			480	(510)	
A—38	105	280	420		480	530	(555)
A—42	115	285	(410)	460	480	(520)	
A—45	120	260	(420)	435	480	530	550
A—49	120	305	390		480	500	
A—52	130	315			480	510	
A—73	125	320				530	
Av.:	126°	297°	(409°)	445°	485°	519°	558°

Values in parenthesis indicate very weak effects.

Table 4 shows first of all peaks characteristic of the organic material of the samples and peaks belonging to the decomposition of carbonates at higher temperatures are neglected. It is to mention that in derivatograms (DTA curve) of some samples fairly intensive exothermic peaks appear in closest neighbourhood of the endothermic peak of carbonate decomposition as shown in *Fig. 2, C.*

Samples of type "*A*" and "*C*" represent the extreme types whereas samples of "*B*" type may be considered as intermediate ones.

Comparing the DTA curves of samples of the "*A*" and "*C*" type the differences at higher temperatures are especially striking. The DTA curves of samples of the "*C*" type show expressed exothermic peaks at 760–860° C (average 796°) and 840–900° C (average 865° C) immediately before and behind the carbonate endo-

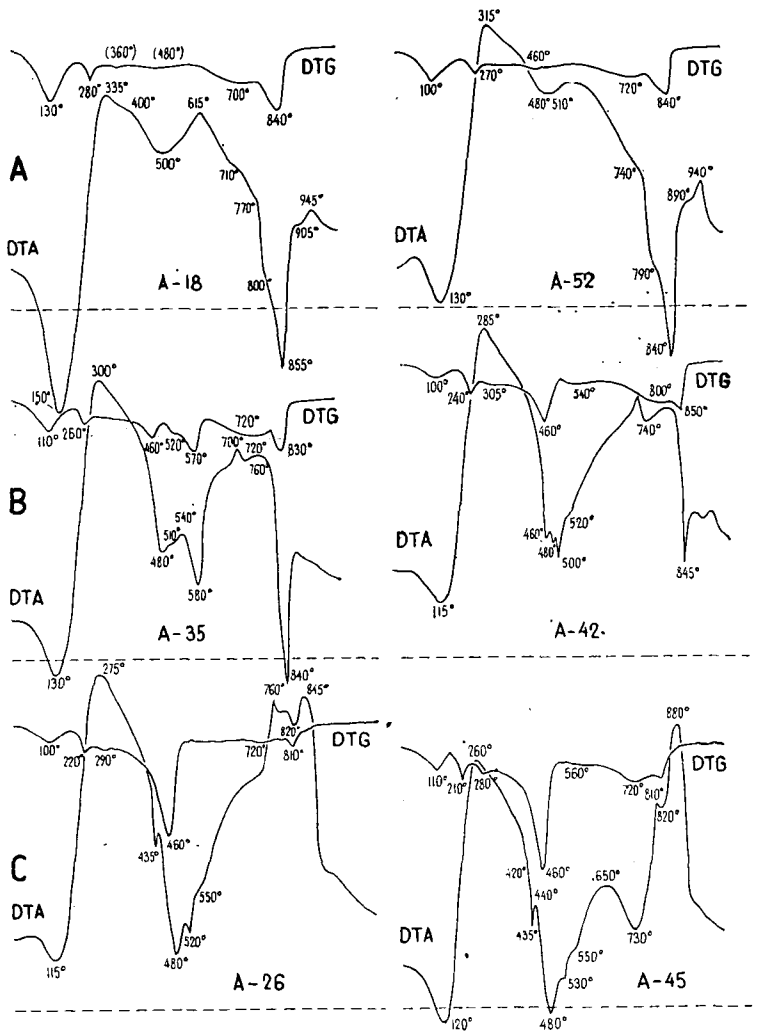

Fig. 2. DTG and DTA curve of some samples from boring Put-7. *A:* A—18 and A—52, *B:* A—35 and A—42, *C:* A—26 and A—45.

thermic peak at 815°–865° C (average 830). The DTA curves of samples of the "*A*" type don't show any exothermic effects within the temperature intervals mentioned. In the DTA curves of samples belonging to the intermediate type "*B*" an exothermic peak at about 700–800° C can also be established, however, it always precedes the well developed carbonate endothermic peak. The main types naturally cannot be sharply delimited. The character of the DTA curves is determined by the changing amount of the organic material as well as by the changing ratio among the amount of organic material, phyllosilicates and carbonates and by the interaction between organic and inorganic components. The organic material content is the highest in samples of the "*C*" type and the lowest in samples of the "*A*" type.

As to the DTG curves, the three maxima following the first one (loss of water) denote the decomposition of the organic material. The first of them from 180° C to 280° C is generally small, however, sharp. The maximum from 280° C to 360° C is hardly or not observable in the curves of most of the samples. The last maximum from 450° C to 480° C, belonging to the decomposition of the organic material shows the relatively lowest variation.

In the DTA curves the temperature of the first endothermic peak varies between 105° C and 155° C, on average 126° C. It is immediately followed by a strong exothermic peak from 260° C to 335° C. In the descending part of this exothermic effect further one or two smaller peaks can be observed in the DTA curve of some samples. The last peak (endothermic) belonging to the decomposition of the organic material appears within the range 480° C to 500° C.

This last peak both in the DTA and DTG curves shows a fairly small variation within a narrow temperature interval of 20°–30° C independent of the depth, the rock type as observed in the case of all samples and merely its intensity is changing depending upon the amount of the organic material.

The first and strong exothermic peak temperature, however, shows a greater variation both in the DTA and DTG curves. Its temperature is the lower the higher is the organic material content.

According to the connection shown by *Fig. 3* the samples investigated may be divided into three groups. Samples A – 14, – 15, – 18, – 32, – 33, – 52, – 73 belong to group "*A*"; samples A – 35, – 38, – 42, – 49 to group "*B*" and group "*C*" includes the samples A – 24, – 26, – 27, – 28, – 45 with the highest organic carbon content.

The increase of the peak temperature in connection with the decrease of the organic carbon content is the consequence of the fact that the decrease of the amount of organic material means at the same time the increase of the amount of inorganic

TABLE 5

Mean values of the first exothermic DTA peak, the total organic carbon and the soluble organic material of groups A, B and C

Group	Temperature of the first exothermic DTA peak	ΣC_{org} %	Total soluble organic material %
A	323° C	9,53	1,64
B	292	17,10	3,83
C	265	36,56	5,76

The connection of the mean value of the first exothermic peak temperature with that of the ΣC_{org} in the single groups is also shown in *Fig. 3* (—*Δ*—*Δ*—).

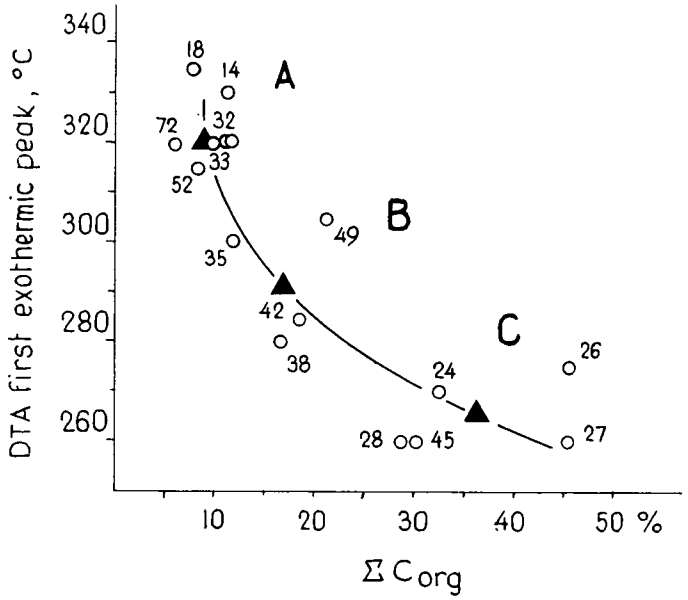

Fig. 3. Connection of the temperature of the first exothermic DTA peak with the total organic carbon content of the samples.

components (clay minerals, carbonates) and the thermal decomposition of kerogen particles surrounded by clay minerals and carbonates is retarded, whereas, in presence of organic material of greater amount the ratio of organic material/inorganic components is shifted towards the organic material and the thermal decomposition proceeds already at lower temperatures.

Under similar conditions the derivatograms of a Green River oil shale sample of Eocene age (Tosco Mine, Rio Blanco County, Colorado, USA) and of a torbanite sample (Newness, near Glen Davis, N. S. Wales, Australia) of Permian age were also recorded as shown in *Fig. 4*.

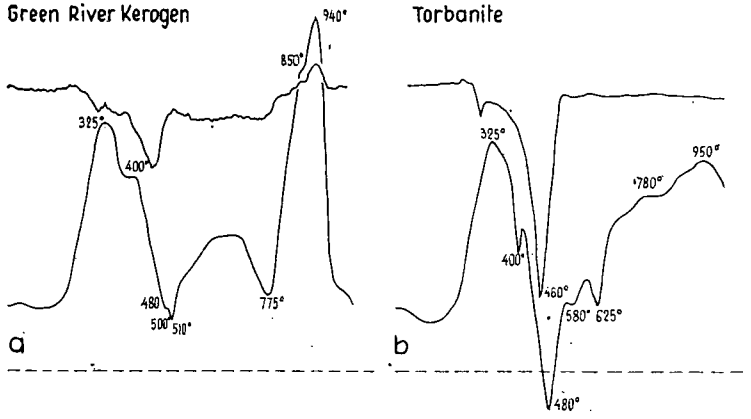

Fig. 4. DTG and DTA curves of a Green River oil shale (a) and torbanite from Australia (b).

In the DTG curve of the Green River oil shale maxima can be observed at 325° C, 360° C and the most intensive maximum at 460° C. The corresponding peaks in the DTA curve are at 325° C and 400° C as well as the peak triplet at 480–500–510° C. This endothermic peak is coupled with the highest loss of weight.

In the DTG curve of the torbanite also the maximum at 460° C is dominant and correspondingly the endothermic DTA peak at 480° C is also dominant and is incident to the greatest loss of weight (49.6 per cent) as registered in the TG curve within the temperature limits from 380° C to 520° C. The temperature of the first exothermic peak in the DTA curve is also 325° C as in the case of the Green River oil shale. On the descending side of this peak a further smaller but definite peak can be observed.

The derivatograms of these two oil shale samples show a very close similarity to those of the Pula oil shale samples rich in organic material. The similarity is especially striking between the Green River oil shale and the samples of Pula belonging to group "*C*", the DTA curves of both show a strong exothermic peak at higher temperatures indicating the final combustion of the fixed carbon. Similarity appears also in that respect that the temperatures (on average) both in the DTG and DTA curves of the comparative samples and the Pula samples, indicating the process of the greatest loss of weight, get on well together (DTG: 461°C, DTA: 485°C).

Differences between the derivatograms of the comparative oil shale samples and those of the samples from boring Put-7 appear in the temperature of the first exothermic peak. The first sharp maximum appears at 280°–290°C in the DTG curve of the comparative samples, whereas, this peak temperature can be observed in the DTG curves of samples from Pula, having a lower organic material content. The temperature of this first maximum in the case of samples rich in organic material (group "*C*'') varies from 180° C to 220° C. Consequently the temperature of the first exothermic DTA peak is lower in the case of samples of Pula varying from 260° to 335° C (on average 297° C) instead of 325° C observed at the comparative samples. The peak temperature varies from 260° C to 275° C in the case of samples rich in organic material (group "*C*'').

The differences mentioned may be interpreted that the oil shale of Pula contains more organic components volatile at a lower temperature and since the temperature of the first characteristic peak should be shifted toward higher temperatures by the increase of the grade of coalification, hence, the alginite from Pula younger than the comparative samples, and originating from shallow depth (10–30 m from the surface) are still in the initial stage of their evolution.

It was established that the temperature of maxima, indicating the largest loss of weight both in the DTG and DTA curves of the samples from Pula and the comparative samples, agree well (461° C and 485° C on average, respectively, at samples from Pula and 460° C and 480° C, respectively, in the case of the Green River oil shale and the torbanite). This fact may be interpreted that this peak temperature is already independent of the age, the type and amount of the organic material, the type of the sediment, thus, the investigation of processes taking place at lower temperature, and indicated by DTG and DTA peaks in the lower temperature interval is more essential from the point of view of the evaluation of the original type and thermal transformation of the kerogen of the sediments. The characteristic peaks in the derivatogram are influenced by the original state of the kerogen at lower temperatures, however, after the decomposition processes having taken place by increasing temperature, the originally existing differences among kerogens are

disappeared so, in consequence nearly the same temperature values can be observed as shown above.

D. R. WILLIAMSON [1964] gives an overview on the results of investigation regarding the peculiarities and beneficiation of oil shales. From the viewpoint of the present investigations the statement seems to be essential that no kerogen is converted to soluble material below 325° C [ROBINSON and HUBBARD see WILLIAMSON] and that according to DULHUNTY [see WILLIAMSON] a sharp increase in the rate of production of soluble bitumen occurs at 360° C, and the rate decreases sharply at 380° C. According to GUTHRIE [see WILLIAMSON] heavy oil vapors and gas begin to be formed and distilled and at about 500–600° C the production of condensable oil vapors ceases.

Considering the samples investigated, rich in organic material (group "*C*") the greatest loss of weight starts at 330–345° C (on average 340° C) and ends at 505–550° C (on average 527° C). The corresponding DTG and DTA maxima are 460° C and 485° C, respectively. The highest loss of weight can be observed from 380° C to 530° C (the loss of weight is 49.6%) at the torbanite and from 380° C to 520° C (10.8%) in the case of the Green River oil shale.

The highest loss of weight and the corresponding temperature limits in the case of samples from Pula (group "*C*", rich in organic material) are as follow:

Sample	Step in the TG curve	Loss of weight %
A—24	from 335 °C to 540 °C	26,5
A—26	340 °C 550 °C	31,2
A—27	300 °C 520 °C	38,7
A—28	330 °C 520 °C	29,7
A—45	330 °C 535 °C	24,8

Derivatographic investigation of kerogen concentrates

First the soluble organic material of the samples was extracted and the kerogen of 11 samples by physical method and that of 5 samples by chemical method was concentrated. Kerogen concentrates obtained by physical method have higher ash (inorganic component) content, however, no change in the structure and composition of the original kerogen occurs. Concentrates obtained by chemical method have lower ash content, however, the kerogen undergoes some changes. Regarding the isolation of the kerogen of the Hungarian oil shales we refer to the paper of M. HETÉNYI and I. VARSÁNYI [1976].

For isolation by physical method the kerogen was treated first by chloroform and the further concentration of the fraction rich in organic material was carried out by treatment (fractionation) with methanol-chloroform mixtures of successively decreasing specific gravity.

Isolating the kerogen by chemical method, first the carbonates were removed and after that the chloride-free washed sample was homogenized by zinc chloride solution (sp. gr. 1.92) and centrifuged. The fractions were separated and the separation of the lowermost fraction was repeated. Finally the corresponding fractions were united washed and dried at 60° C. The further treatment was carried out according to LÜCK [1969] using acid treatment with hydrofluoric acid and alternating

treatment with dilute hydrochloride acid and ammonium hydroxide and finished the isolation with washing and drying the concentrate.

The derivatograms of the concentrates were recorded under similar conditions as those of the original samples only a special sample holder was used which results in a better separation of the peaks.

Table 6 shows the values of ignition residue of the original samples and the corresponding concentrates, respectively in the case of samples isolated by physical and chemical method. The samples were heated to 1000° C.

TABLE 6

Ignition residue of original samples and kerogen concentrates

Sample	Ignition residue %			
	original sample		concentrate	
A—14	61,41	concentrated by chemical	6,70	
A—15	63,29	method	7,20	
A—18	62,94		1,00	4.12
A—52	61,82		0,00	
A—73	57,86		2,70	
A—24	44,12		7,67	
A—26	38,75	concentrated by physical	8,75	
A—27	33,96	method	8,50	
A—28	42,17		7,86	
A—32	62,15		8,75	
A—33	60,15		7,20	7,14
A—35	54,96		2,00	
A—38	41,05		8,00	
A—42	51,97		6,88	
A—45	43,37		7,40	
A—49	52,20		12,67	

The characteristic peaks in the DTG and DTA curves of the concentrates are comprised in Table 7.

According to the thermogravimetric measurements the very first part of the TG curve ends on average at 170° C and is accompanied with a very low loss of weight, followed by two more expressed sections in the case of kerogens physically concentrated. Thus, the second and significant step begins at 170° C and ends at 376° C on average and is accompanied with a loss of weight of 35.52 per cent on average. The third considerable step begins at 376° C and ends at 636° C and the average loss of weight is 53.19 per cent. Within the two large steps smaller ones can be observed.

The first step in the TG curve of the concentrates obtained by chemical method ends at 155° C and represents 5.33 per cent loss of weight (on average), the second intensive step ends at 373° C and is accompanied by a loss of weight of 30.89 per cent. The last step is terminated at 615° C on average leading to a loss of weight of 58.08 per cent. The combustion processes stop at that temperature and in the curves no more changes can be observed.

Fig. 5 shows the DTA and DTG curves constructed on the basis of the mean values of temperature and loss of weight, respectively, both for kerogens physically and chemically concentrated.

TABLE 7

The characteristic peaks in the DTG and DTA curves of the concentrates

a) Peak temperatures in the DTA curve (°C)

Kerogen concentrated by physical method

	I	II	III	IV	V	VI	VII	
A—24	225	300		375	440	(500)		
A—26	250	(290)	(340)	380	450	(510)	(580)	
A—27	205	285	(350)	385	450	(520)		
A—28	230	300	(340)	370	450			
A—32	235	290	(350)	385		(500)		
A—33	235	300	(340)	375		(480)	(555)	
A—35	220	300	(350)	385		(510)		(605)
A—38	230	310	(355)	390	460		(570)	(595)
A—42	230	290	(340)	375		(485)	(580)	(650)
A—45	215	295	(340)	365	435	(500)	(580)	
A—49	260	(290)	(350)	380		(490)		
Average:	230°	295°	(345°)	378°	460°	(499°)	(573°)	(616°)

Kerogen concentrated by chemical method

A—14		285	(335)	385	465	(510)	(580)	
A—15		300	(340)	380	450			
A—18		320	(355)	400	425	490	570	
A—52		310	(350)	370	400	440	550	
A—73		300	(360)	380	420	450	555	
Average:		303°	(348°)	383°	432°	472°	563°	

b) Temperature of maxima in the DTG curve

Kerogen concentrates by physical method

	I	II	III	IV	V	VI	VII	VIII	IX	X
A—24	220		300	340		(440)		500		
A—26	230		310	340		(450)		470		580
A—27	180		285	350						620
A—28	220		300	370		(450)		485		
A—32	220		300	340		(460)		500		580
A—33	230		300	340		(440)		480		560
A—35	220		300	340		(440)		530		615
A—38	220		300	355				500		595
A—42	210		300	350		(450)		500		630
A—45	200		295	340		(440)		500		580
A—49	240		320	350		(450)		500		560

Average:	217°	301°	347°	(447°)	496°	591°
			Kerogen concentrates by chemical method			
A—14	265		(320)	480	520	
A—15	280		(340)	470	540	
A—18	300		(355)	440	520	600
A—52	280		400	465	515	570
A—72	270		(350) 420	440	515	
Average:	279°		(341°) 410°	459°	522°	585°

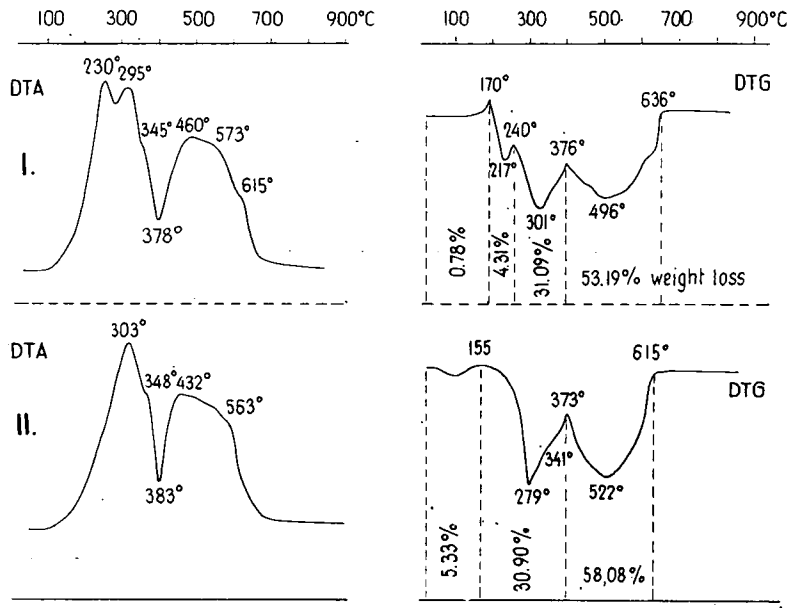

Fig. 5. DTA and DTG curves constructed on the basis of the mean value of experimental data
I: concentrate by physical method;
II: concentrate by chemical method.

From the data in Table 7 and the temperature limits of the steps in the TG curves as well as the loss of weights accompanied with them, the following conclusions may be drawn regarding the kerogens isolated from oil shale from Pula:

(i) Kerogens isolated by the two ways mentioned are similar in that respect that heating them on the air the combustion of the organic material is divided into two large sections (steps). The first one including 2 or 3 smaller steps, too, terminates nearly at the same temperature in case both of kerogens concentrated physically and chemically (376° C and 373° C). Similarity can also be established so far as the first reactions result in expressed, sharp peaks in the DTA and DTG curves, whereas, the second effect is broad and vaulted without any sharp peaks. The first represents the lower and the second section the larger loss of weight. Within the second section smaller but uncertain steps (peaks) may be observed. The combustion of the kerogen concentrated physically stops at 636° C and at 615° C in the case of kerogen isolated by chemical method. Summarizing the partial losses of weights during the first reac-

tions taking place to 376° C, nearly the same values are obtained: 36.18 per cent in the case of kerogen concentrated by physical method and 36.23 per cent at the kerogens isolated by chemical way.

(ii) Difference between the kerogens concentrated in the two manners mentioned appears definitely in processes occuring at lover temperatures. The TG curve of kerogens physically concentrated hardly shows any loss of weight to 100–120° C, even a very small increase of weight can be observed up to 160–170° C which may be interpreted as an oxidation process. The TG curve of kerogens isolated chemically starts with an expressed step representing a loss of weight larger than that observed in the TG curve of kerogens physically isolated.

The DTA curve of kerogens concentrated in physical manner is characterized by an intensive, sharp exothermic peak from 205° C to 260° C (on average 230° C) which is totally lacking in the DTA curve of kerogens concentrated by the other way. In the DTA curve of the latters the first sharp exothermic peak appears at 303° C coinciding with the second exothermic peak of kerogens physically isolated, occuring at 295° C. Consequently, a sharp maximum appears in the DTG curve of kerogens concentrated by physical way at 217° C (on average), which is lacking in the DTG curve of the kerogens chemically concentrated. This maximum corresponds to the first exothermic DTA peak.

On the second broad exothermic peak somewhat higher temperature values can be observed in case of kerogens concentrated by physical method. The character of this second broad and vaulted section is very similar in the case of kerogens isolated by the two different methods.

(iii) It may be supposed that during the concentration by chemical method some structural changes occur (rupture of bonds, oxidation etc.), whereas the original structure of kerogen remains intact by isolation physically, and the decomposition of kerogen starts only under the influence of heat. E.g. we can consider decarboxylation, followed by oxidation of the terminal methyl groups to carboxyl and with the repetition of this process similar to the decomposition mechanism described by B. LÓRÁNT [1972] in his study on the thermal decomposition of fatty acids.

The aim of further investigations is to establish the connections between the possible steps of decomposition of the kerogen from Pula and the effects observable in the derivatograms.

(iv) It seems that the type of kerogens from the different levels of the oil shale occurrence at Pula can be considered as nearly identical, because the kerogens from different layers of the occurrence gave very similar derivatograms *(Fig. 6)*. The bulk of the organic material is built of *Botryococcus braunii* and the deviations among the derivatograms are due to the presence of other organic material (pollens, coalified plant remnants etc.) besides the *Botryococcus*.

(v) The effects appearing in the derivatogramms of the original samples at lower temperatures, and accompanied presumably by the decomposition of more volatile organic material are characteristic, however, they are influenced by the inorganic components, hence, the DTA curves of the original samples represent the effects of this interaction. In the DTA curve of some samples (e.g. Group "*C*") a fairly strong exothermic peak appears at higher temperature, whereas, the DTA curve of kerogen concentrated from the same samples did not show any similar effect, the combustion process is finished at 620–640° C.

Since the particles of the organic material are included in clay minerals and carbonates, respectively, and the bonding between organic and inorganic components apparently can be strong [WILLIAMSON, 1964], the combustion of the organic material

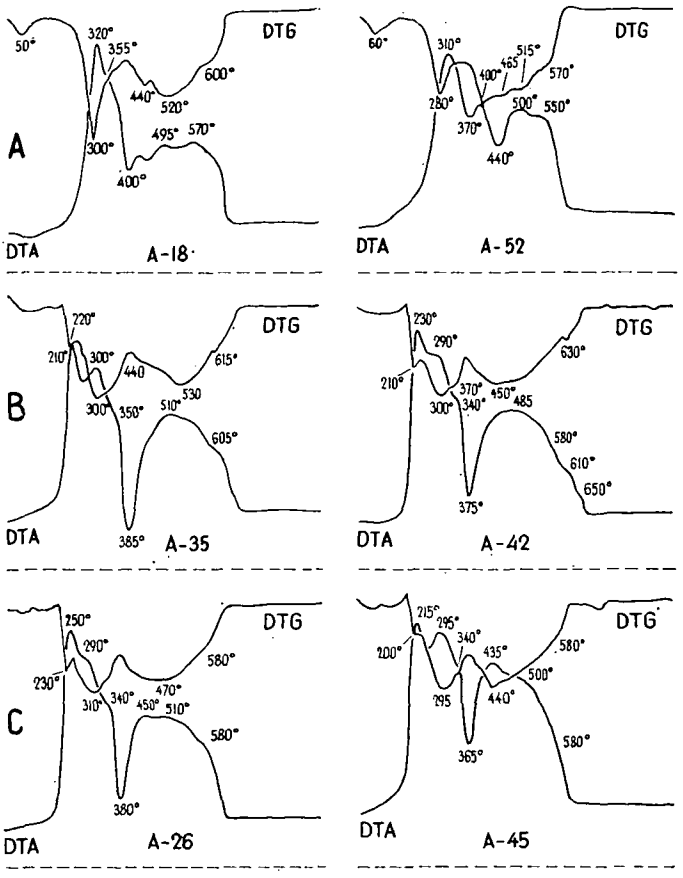

Fig. 6. DTA and DTG curves of kerogen concentrates isolated from samples denoted

at lower temperatures is retarded. Thus, after the decomposition of the organic material, what is finished very soon above 460° C in the case of the original samples according to the DTA curves, the final combustion of the fixed carbon is shifted towards higher temperatures accompanied by the decomposition of the phyllosilicates and carbonates, respectively. It is especially striking in the case of samples rich in organic material, however, it may be observed to a lesser extent also in case of samples of medium organic material content.

IR-SPECTRA OF KEROGEN CONCENTRATES

Comparison of IR-spectra of kerogens concentrated by physical and chemical methods

IR-spectra of the kerogen samples were recorded by Zeiss UR–20 spectrophotometer from 4000 to 600 cm^{-1} wave number, in KBr pellet (KBr: 800 mg, sample 5.0 mg). *Fig. 7* shows well the differences between the IR-spectra of kerogens physically *(kerogen I)* and chemically *(kerogen II)* concentrated.

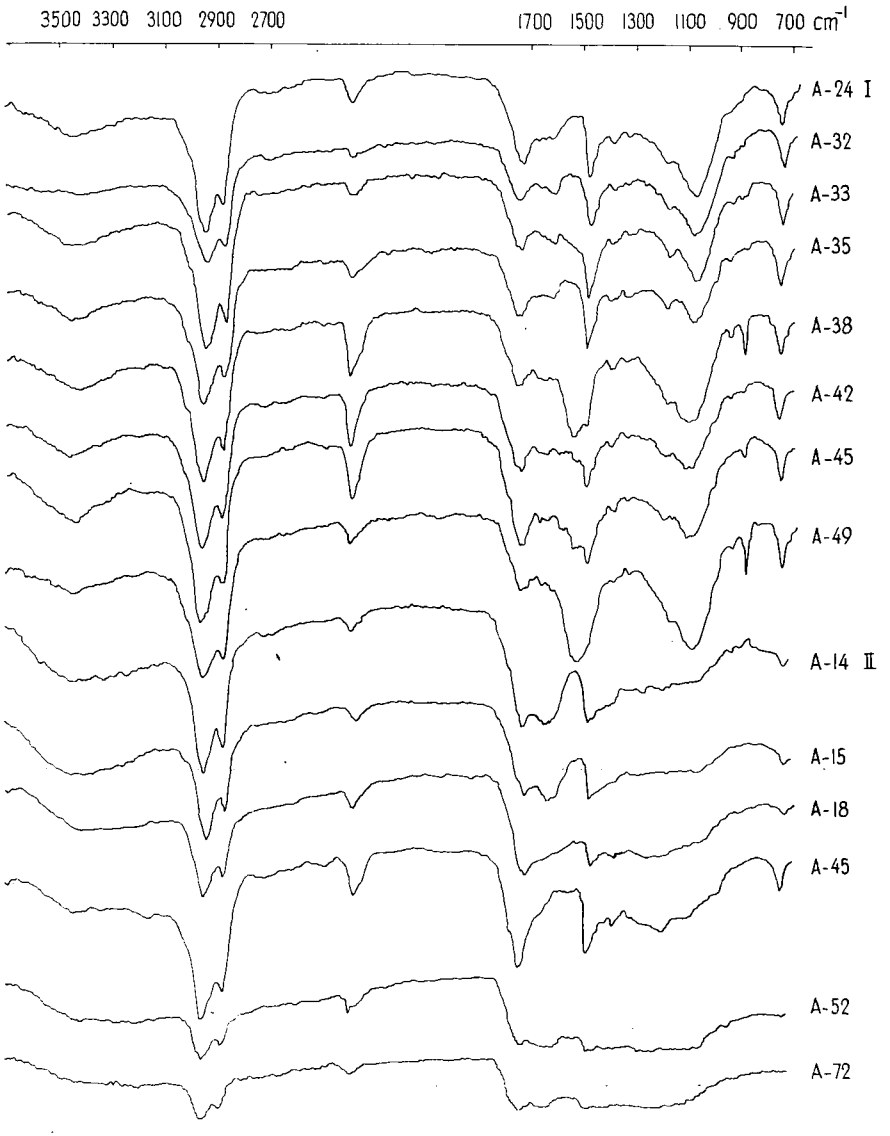

Fig. 7. IR-spectra of the kerogen samples.

Considering the characteristic absorption bands, the following can be established:

(i) In the case of *kerogen I* concentrates, the broad absorption band at 3700–3200 cm^{-1} may be assigned to OH streching vibration originated from alcoholic and/or phenolic OH-groups, and moisture, respectively. The latter may be suggested from the symmetric shape of the curve. This absorption band of *kerogen II* concentrates at 3700–3100 cm^{-1} is broader and non symmetric. The weak band at 1400–1350 cm^{-1}, hydroxyl in-plane deformation vibration also points to the presence of OH-groups.

TABLE 8

Absorption bands in the IR-spectra of kerogen I and kerogen II samples

Kerogen I		*Kerogen II*	
Wave number cm^{-1}	Intensity	Wave number cm^{-1}	Intensity
3700—3200	m, (broad)	3700—3100	m, (broad)
2950	vs	2950	vs
2870	s	2870	s
2720—2680	vw	2720—2680	vw
2450	s	2450	m
1750	s	1750	s
1720	s	1720	s
1600	w	1600	m
1550	w	—	
1480	s	1480	m
1400—1350	w	1400	w
1180	m	—	
1080	s	—	
950	vw	—	
920	vw	—	
720	m	720	vw

vs: very strong; s: strong; m: middle; w: weak; vw: very weak

(ii) The most intensive bands of the spectra at 2950 and 2870 cm^{-1}, respectively, can be assigned to the symmetric and asymmetric streching vibration of methyl and methylen groups. The overlapping of 2–2 bands point to the complex structure of the samples investigated.

In spectra of the *kerogen I* concentrate the intensity of these bands is stronger than those of the *kerogen II* concentrate and it suggests the assumption that the amount of methyl and methylen groups decreased due to oxidation during the chemical treatment. A similar phenomenon can be observed also in the case of the band at 1480 cm^{-1} assigned to the in-plane deformation vibration of methylen groups. The *xy*-deformation vibrations and the in-plane deformation vibrations of methyl and methylen groups coincide with the in-plane deformation vibrations of the OH-group (1400–1350 cm^{-1}).

The band at 720 cm^{-1} may be explained as an in-plane deformation vibrations of groups containing side-chains with four or more carbon atoms. The intensity of this band is stronger in the spectra of the *kerogen I* concentrate.

(iii) The bands at 1750 and 1720 cm^{-1} can be originated from the symmetric and asymmetric vibrations of carbonyl-groups. In spectra of *kerogen II* concentrate the intensities of these bands are nearly equal with those of the methyl and methylen groups at 2950–2870 cm^{-1}, whereas, the intensity of carbonyl groups is decreased in the case of *kerogen I* concentrate.

(iv) The absorption band at 1600 cm^{-1} can be assigned to the aromatic and/or unsaturated parts of the kerogen molecule.

(v) There are some bands (1180, 1080, 950, 920 cm^{-1}) observed only in spectra of the *kerogen I* concentrate, which may be originated partly from aliphatic and

aromatic esthers and aliphatic ethers partly from the mineral constituents not eliminated by the isolation process.

At 2450 cm^{-1} a strong band in the case of *kerogen I*, and a middle strong in the case of *kerogen II* could be observed. It may be assumed that it is a combination band of C—O streching vibration and OH in-plane deformation vibration.

Changes in the IR-spectra of kerogen concentrates heated to different temperatures

Portions of sample No. 45 concentrated by physical and chemical method, respectively, were heated to given temperatures in the Derivatograph and the IR-spectra of products obtained at different temperatures were recorded by Pye Unicam SP 1100 spectrophotometer. The actual temperature values reached were read from the T curve of the derivatogramme:

Kerogen I: 260°, 310°, 370°, 430° and 575° C,

Kerogen II: 260°, 310°, 395°, 450° and 600° C.

The increase of the temperature resulted changes in the IR-spectra as follows:

(i) The intensity of OH absorption band gradually decreases. At 450° C it can already not be observed in the case of *kerogen II* samples, whereas, in the case of *kerogen I* samples in spite of a strong decreasing tendency, it can be observed still at the highest temperature applied. This fact may be interpreted that the decomposition of the structure of the phyllosilicates (clay minerals) present is not yet completed at this temperature. The band 900–850 cm^{-1} appeared only in the spectra of physically concentrated kerogens and does not show any change during the increase of the temperature.

(ii) The intensity of bands at 2950–2870 cm^{-1} decreases by increasing temperature in both set of kerogen concentrates, and it shows minimum already at 370–395° C. A similar decrease of intensity can be observed in the case of bands at 1480–1460 cm^{-1} belonging to the methyl-methylen groups.

The intensity of the band at 740–720 cm^{-1} shows a decrease parallel to that of the methyl and methylen groups.

(iii) The intensity of bands at 1750–1720 cm^{-1} (C=O of carbonyl and carboxyl groups) first increases by increasing temperature, however, from 370–395° C a decrease of the intensity can be stated.

(iv) The absorption band at 1610–1600 cm^{-1} shows an increasing tendency in both set of kerogen concentrates and it is the strongest band of the spectrum at 370–395° C.

(v) The intensities of bands at 1140–1000 cm^{-1} observed only in the spectrum of the *kerogen I* samples remained unchanged during the heat treatment, thus, they can be originated from inorganic components.

The change of the intensity ratios of absorption bands assigned to the different groups, in function of the temperature, was also considered.

The E_{2900}/E_{1600} ratio shows a sharp decrease in both set of kerogen concentrates up to 370–395° C, and from this temperature the curve is flattened showing only a slight decrease.

The change of the E_{2900}/E_{1700} intensity ratio is very similar to the former.

Comparing the change of intensity ratios of E_{1700}/E_{1600} in the spectra of the *kerogen I* and the *kerogen II* concentrates, it can be established that the curve for the *kerogen I* exhibits a more or less uniform decrease, especially up to 420–430° C,

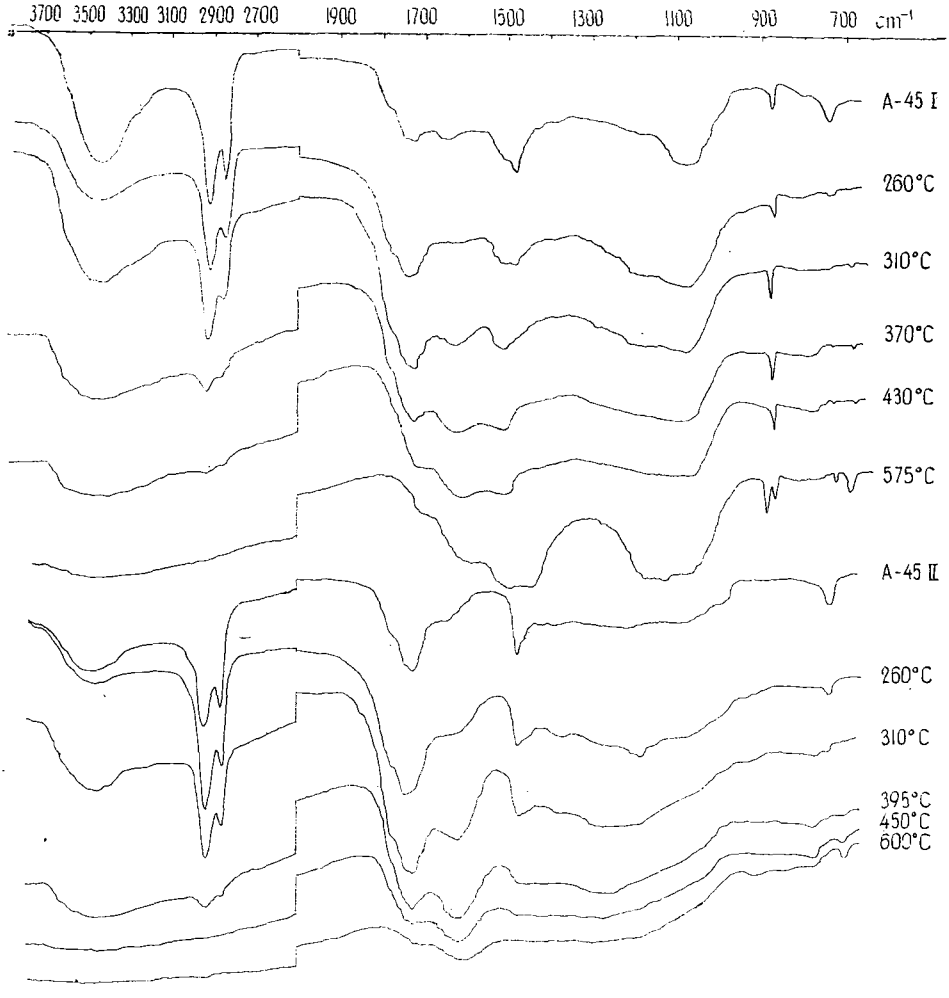

Fig. 8. IR-spectra of kerogen concentrates heated to different temperatures.

and the first section of the curve for the *kerogen II* is somewhat steeper, however, between 300 and 430° C the two curves are nearly parallel to each other.

In comparing the intensity ratios observed in the case of samples heated to 260° C, it can be established that the ratios E_{2900}/E_{1600} and E_{1700}/E_{1600} are higher in the spectra of kerogen chemically concentrated, whereas, in that of isolated by physical method the E_{2900}/E_{1700} ratio shows a higher value. It may be assumed that the methyl-methylen groups remained rather unattacked during the isolation by physical method, whereas, the isolation by chemical processes resulted in at least partial oxidation. The aromatic part of the structure of the kerogen molecule seemingly remained unchanged either using the physical isolation or the chemical one. The amount of aromatic part shows merely a relative increase due to the gradual decomposition of other functional groups under the influence of heat.

The H/C atomic ratio exhibits a decrease in function of the temperature as it is seen in *Fig. 9*, and no essential difference occurs between kerogen samples isolated by physical and chemical method, respectively.

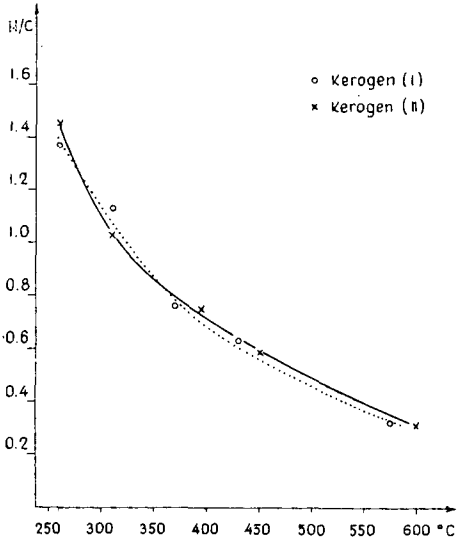

Fig. 9. Change of the H/C atomic ratio of *kerogen I* and *II* under the influence of heat.

The analytical determination of the amount of functional groups in the kerogen samples heated to different temperatures and the study of the pyrolysis in nitrogen stream presumably will lead to a more detailed picture.

CONCLUSIONS

Comparing the change of the intensity ratios of different IR-absorption bands in function of the temperature with the experiences of the derivatographic examinations the following conclusions may be drawn:

(i) The validity of the decomposition mechanism according to LÓRÁNT is very probable regarding the methyl-methylen as well as the carboxyl groups.

(ii) The first section of the DTG curve up to 376° C may be characterized by the gradual oxidation of methyl-methylen groups and decomposition of the carboxyl groups, in accordance with the character of the change of E_{2900}/E_{1700} and E_{2900}/E_{1600} ratios exhibiting a fairly steep decrease up to 370–395° C.

(iii) The intensity of the absorption band assigned to aromatic part of the structure is fairly weak and its change under the influence of heat is far not so expressed as the change of the intensities of the bands belonging to the methyl-methylen and carbonyl-carboxyl groups.

(iv) The methyl-methylen bands can be observed in spectra of kerogens heated to 395° C but they are lacking already in the spectra of samples heated to 450° C. At the same time, the band belonging to the carbonyl-carboxyl groups, weak but well observable at 395° C, can be detected also at 600° C. Furthermore, the intensity of the aromatic band shows an increasing tendency up to 395° C, then between 450–600° C decreases, but even at 600° C remains well observable.

These facts suggest that the second section of the DTG curve (from 376° to 636° C in the case of the *kerogen I* concentrate and from 373° to 615° C in that of the *kerogen II* sample, on average) represents the decomposition of the carbonyl-carboxyl groups not yet decomposed and dominantly that of the aromatic structures. In the first section of the DTG curve (and similarly, of the DTA curve) well defined, sharp peaks appear, the rate of reactions taken place within this temperature interval is faster than that of the reactions in the second DTG (and DTA) section. The second section of the DTG curve extends over a somewhat larger temperature interval than the first one, and individual peaks can not be distinguished in it, they are united in a broad maximum. These facts are in accordance with the rate of changes of intensity ratios of characteristic absorption bands.

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IAGOD COMMISSION ON MANGANESE IGCP PROJECT NO. 111: GENESIS OF MANGANESE ORE DEPOSITS

NOTES AND NEWS

ACHIEVEMENT AND SCIENTIFIC SIGNIFICANCE OF THE IGCP PROJECT No. 111: GENESIS OF MANGANESE ORE DEPOSITS

by
GY. GRASSELLY
Project leader

The 2nd International Symposium on the Geology and Geochemistry of Manganese (25th IGC, 1976, Sydney) is to be considered as the start and the first result of the Project based on the activity of the IAGOD Commission on Manganese. The Monograph on the Geology and Geochemistry to be published (in English) in three volumes in 1978 can also be considered as a significant progress in the Project.

The lectures read at the Symposium and the papers to be published in the Monograph review not only the results and the progress in different fields of manganese research, moreover, they outline the directions of research, where for the sake of a successful progress a wide international co-operation is possible and necessary. The role of the Project is to promote and extend the active connections so far existing, drawing into the common work further institutions, groups and individuals interested in this field.

The progress made in different directions of manganese research could hardly be enumerated or even roughly outlined; it is only possible to point to the most interesting and promising topics with reference to works and authors already having a closer or looser connection to the Project.

Reviewing the huge scope of manganese research, it seems that special attention is focussed on the geology and genesis of manganese ore deposits on the continents, and on the origin and distribution of manganese and associated metals on the bottom of recent basins. Naturally, all the questions to be solved in these fields and those in general in the mineralogy and geochemistry of manganese, are closely interwoven.

Studies on the geology and genesis of manganese ore deposits on the continents are characterized by a multidisciplinary approach of the problems in their full complexity, taking into consideration the mineralogical, geochemical and petrological aspects, with special stress on the determination of the nature of the basins and the environments of deposition.

Research work carried out recently all over the world completed or modified our knowledge concerning the manganese ore deposits of different origin and age only to mention the major deposits as the huge manganese ore deposits in the Soviet Union, the Indian manganese ores or manganese deposits in Japan, the genetically

very interesting manganese deposits in Brazil, Australia, Gabon, Ghana, which are very promising from the viewpoint of further research work.

Detailed studies of major manganese deposits, the studies of the protores and their weathering products as well as the investigations of the effect of metamorphism and the metamorphic mineral assemblages etc., considerably enlarged our horizon in this respect.

The thorough consideration of the conclusions of these extensive researches propounds, however, questions and ambiguities worthy of further study in the Project. Thus, it would be useful to study the genetical processes of phosphorite-manganese oxide associations for the sake of a better understanding of the behaviour of associated phosphorous (apart from the formation of apatite) in manganiferous sediments during metamorphism. Furthermore, there is no satisfactory explanation for the fact that important deposits of braunite and manganese oxide-silicate rocks are present in association with carbonaceous rocks or that graphite is present in an oxide-silicate assemblage (Guiana Shield, South America and Marau, Brazil, resp.). Likewise, a very interesting question: how the presence of high-temperature manganese oxides of lower valences as hausmannite, jacobsite, braunite, bixbyite may be interpreted in orebodies without any sign of metamorphism, as it was raised by Professor SUPRIYA ROY, Leader of Panel I.

Very active and successful researches are presented also in the study of distribution of manganese and associated metals on the bottom of recent basins including the lakes, deep-seas and shallow-seas, in close conncetion with studies of problems of mineralogy and geochemistry of manganese and associated metals.

Wide-ranging investigations were carried out to study the submarine volcanism at spreading centers as a source for metalliferous deposits. Results suggest that the ferromanganoan active ridge sediments are only to a small extent due to the leaching of oceanic rocks or deposition from ordinary sea water, and a hypothesis was rendered probable by a lot of data that emanations of deep-seated origin, probably rich in CO_2 are carriers of Fe, Mn and other elements.

The connection between mineralization and the subduction processes at convergent plate margins is also one of the thoroughly investigated problems.

The Transatlantic Geotraverse Project of the National Oceanic and Atmospheric Administration discovered the TAG Hydrothermal Field in the Mid-Atlantic Ridge where hot springs enriched in various metals are apparently discharging from the sea floor and depositing a solid layer of nearly pure manganese oxide. It was established that types of metal deposits occurring in oceanic crust formed by sea floor spreading occur only along oceanic ridges where conditions are favourable; the distribution of localities is presently unknown.

It was demonstrated by a regional geochemical reconnaisance survey of marine sediments in the Southwest Pacific that the composition of these sediments shows a wide variability, and, in an area associated with a belt of earthquake epicenters, metal values higher than average were observed.

The progress raises at the same time the directions of further researches.

The concise programme in general terms, elaborated by Professor DAVID S. CRONAN, Leader of the Subdivision 3b aims to evaluate the influence of submarine volcanism on the composition of manganese nodules and encrustations, particularly its role in determining their regional compositional variability throughout the World Ocean. Therefore, it is proposed to investigate the chemistry of concretions from known volcanic areas in order to define characteristics which can be assigned to submarine volcanic activity. As the correlation part of the work nodules and encrus-

tations from possibly volcanic areas should also be examined and compared with those from the known volcanic areas to see if they show any characteristics which would enable an assessment of volcanic influence on them to be made. Similarly, it would be of importance to investigate the relationship between hydrothermal submarine metalliferous sediments and associated ferromanganese oxide encrustations to see if chemical correlations between them are possible.

The aims of the Subdivision 3c are closely connected with those of the formerly mentioned Subdivision and the CCOP/SOPAC manganese programme. These aims may be summarized according to DR. G. P. GLASBY, Leader of Subdivision 3c as follows: the distribution and geochemistry of manganese nodules throughout the Pacific Ocean show that the most important problem to be tested is the role of biological productivity and transport in controlling the abundance and metal contents of manganese nodules. Areas of low metal content nodules in the Southwest Pacific associated with low biological productivity should be compared and contrasted with areas of high metal content nodules in the equatorial North Pacific associated with high biological productivity in order to asses whether the metal contents of the nodules are diagnostic of the environment of deposition, and in particular, the productivity of the overlying water. There are many interesting problems to be investigated *e.g.* the occurrence of bottom currents and submarine volcanism and the palaeoenvironment in and between fracture zones and the role of these factors in nodule formation; a comparison of manganese nodules and their associated sediments in the North, Equatorial and South Pacific in order to test the symmetry of distribution of carbonate, siliceous ooze and red clay sediments across the Equator. Of particular importance here is the exact relationship between the nodules of highest abundance and Cu-Ni content and the belt of highest productivity in the equatorial zone.

Scientific task and objectives of Subdivision 3d are shortly comprised by DR. PETER A. RONA, Leader of the Subdivision as follows: the objective is to study processes of concentration of manganese and other transition metals at oceanic spreading centers and to determine the distribution of concentrated metal deposits related to oceanic spreading centers. The approach is interdisciplinary, utilizing geological, geochemical, and geophysical methods to develop an understanding of metal deposits as the product of the total system active in oceanic spreading centers. The research covered by this subdivision includes aspects of the geologic setting, crustal structure, tectonics, petrology, thermal regime, oceanography, geochemistry of solid, liquid and gaseous phases, and other subjects relevant to the processes of concentration of metals at oceanic spreading centers and exploration of metal deposits in oceanic crust.

It may be concluded that the basic problems of manganese researches *i.e.* the origin and distribution of manganese are closely connected with the aspects of plate tectonics, and that it would be advisable for the IGCP Project No 111, the NOAA Project, the CCOP/SOPAC Manganese Programme, the IUGS Commission on Marine Geology and for the U.S. Inter-University Manganese Nodule Project to combine their forces to approach the solution of the problems mentioned, equally important from both a scientific and and economic viewpoint.

A number of works dealt in recent years with the origin, conditions of formation, chemical and mineral composition of iron-manganese nodules, concretions, encrustations in shallow water basins, in lakes and in shelf basins. Numerous observations in terrain and results of laboratory experiments suggest that accumulation products are initially formed as amorphous colloidal phases and that selective che-

mosorption and autocatalytic effects play an important role in the formation of the iron-manganese concretions. It was pointed out that the alternation of iron- and manganese-rich layers is the consequence of the seasonal fluctuations in Eh and pH, because the change of the redox conditions in the lake results in a change in the position of the reducing zone in the sediment.

Considerable progress was made in the interpretation of formation mechanism of the deep sea iron-manganese nodules. Some differences may be seen among the different mechanisms suggested which partly may be due to differences in physico-chemical conditions and chemistry of environments, the activity of microorganisms, the bottom current velocity at the site of deposition, etc.

It is generally agreed that initial deposition of hydrous ferric oxide precedes deposition of ferromanganese oxides on different types of nuclei. This hydrous ferric oxide phase was identified by recent investigations both in laboratory products and deep sea iron-manganese nodules as δ'-FeOOH, an unstable modification which may be transformed into goethite.

Explanation of the formation mechanism of iron-manganese nodules is possible only by inorganic processes. It is assumed that the initially precipitated manganous hydroxide (on the active surface of hydrous ferric oxide) may be at least partly oxidized, and precipitation by adsorption of additional Mn^{2+} from sea water may be proceeded. The autocatalytic oxidation of "10 Å manganite" results in a partial or complete loss of the 10 Å basal structure and associated water layers.

It is worth mentioning that a critical review of x-ray diffraction of nodules shows that the actual evidence does not support all of the evaluations and conclusions of the present literature. So, it is proposed that the phase previously named as "10 Å manganite" should be re-named as buserite. Furthermore, the synthetic experiments in laboratory lead to the conclusion that Mn in colloidal solution in waters will be present — at least as the first phase that can be detected — in form of extremely finely dispersed γ-MnOOH. The question may arise whether or not the nodules are formed in a secondary cycle where the initially precipitated γ-MnOOH is mobilized by reduction and reprecipitated in contact with aerobic waters on an active surface of hydrous ferric oxide. This hypothesis shows a close relation in some respect with an other formation mechanism outlined below.

According to an other mechanism, deposition by precipitation of iron-manganese oxides directly from sea water does not play an important role in the formation of iron-manganese nodules, since Fe and Mn is derived from the buried ooze where reduction reactions proceed. Carbonate complexes of iron and manganese migrate upwards into the upper part of the bottom ooze and at the interface of ooze and sea water $Fe(OH)_2$ is formed and oxidation processes — without participation of iron bacteria — lead to the formation of δ'-FeOOH. Contrary to the iron, manganese is wholly or partly precipitated by bacterial activity. Relics of bacteria as *Metallogenium* and *Siderocapsa* were found in the nodules studied.

It seems that some further questions may arise regarding the identification of manganese minerals of the nodules which may be solved by systematic and comprehensive study of formation, composition, structure, and reactions of oxide-hydroxides of manganese and iron under laboratory conditions on the one hand, and in natural environments on the other hand as it is recommended by DR. R. GIOVANOLI, Leader of Panel II, stressing the importance of investigation of processes which lead from Mn^{2+} ion to manganese sediments in order to establish — where this is not already done — which mineral species (solid phases) exist under given conditions and the importance of studies on reactions of laboratory products under controlled

conditions in order to approach step by step the naturally ocurring systems. Problems in identification of manganese oxide minerals are probably due to the poorly crystallized character of the different phases. Consequently, the identification of finely dispersed manganese oxid-hydroxide phases by different methods is also of importance not neglecting the study of how the particle size and shape and other possible interfering factors (intergrowth, order-disorder phenomena, non-stoichiometry) influence the powder diffraction pattern.

Mineralogical studies and geochemical investigations form an integral part of most research project in the entire field, however, in spite of the progress made recently compiling a synopsis of all important criteria for the determination of the most important manganese oxides and hydroxides, there are still problems in nomenclature and recognition of manganese ore minerals, and also in recognition, description and interpretation of textures in ores — as it was raised by Professor RONALD K. SOREM, Leader of Subdivision 3c — which must be resolved before geologists can be sure that they know how others would interpret the features and minerals they discover. Therefore, it is necessary to encourage uniform standards for optical and x-ray, IR, thermoanalytical etc. studies, and to try to establish formats for presenting data so that findings from all parts of the globe can be compared with ease.

A number of works stress the importance of the role of microorganisms in the genesis of manganese accumulations both in the formation of lacustrine ferromanganese ores and deep sea manganese nodules. Manganese fixing bacteria were described in a Cretaceous chert and the extensive photomicroscopy and scanning electron microprobe study suggests that the bacterium *Metallogenium personatum* described in recent lacustrine sediments had had close analogues on the deep sea floor throughout much of geologic time.

The detailed study of the role of microorganisms in accumulation of manganese is very important because the oxidation of Mn^{2+} due to bacterial activity can take place under such physico-chemical conditions wherein it could not be expected merely by chemical reactions, furthermore, the rate of biogenic oxidation of Mn^{2+} catalyzed by microorganisms is several orders higher than that of chemical processes involved in autocatalytic oxidation.

* * *

The start of the IGCP Project No 111, as it was mentioned, can be traced back to the Sydney Symposium – keeping in mind, of course the work, lasting for about one and half a year, resulting in the International Symposium on Manganese, and collecting the material for the Monograph. The start and the preparation were alike really based upon a wide international distribution of work, and we are convinced that the common efforts experienced in the organisation work will also appear in the scientific work based upon an even distribution of the tasks bringing good results. Here mention should be made of the detailed programme to be outlined in the near future, their realisation, furthermore, of research training and technological transfer serving the vital interests of the developing countries.

The Report is based mostly on the contributions submitted to the 2nd International Symposium on the Geology and Geochemistry of Manganese, which represent the latest results in this research field as well as on the preliminary short programme proposals of Leaders of the panels and subdivisions, respectively. The Project leader would like to express his gratitude to all contributed to the compilation of this Report.

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The Project leader expresses his sorrow that because of the restrictions on Bibliography he could not cite many important papers, what the authors will surely understand.

APPENDIX

Measures of IGCP Project No 111 achievement in addition to scientific results

1. The scope of the Project No. 111 includes the study of metalliferous sediments in the recent basins from the Atlantic to the Pacific, and that of the manganese ore deposits on the continents, therefore, the Project can really count on a global intercontinental interest.

The distribution of countries among continents wherefrom contributions were submitted to the 2nd International Symposium on the Geology and Geochemistry of Manganese is as follows: Europe: six countries with 37 contributions; America; two countries with 11 contributions; Asia: three countries with 6 contributions:

* Papers denoted with asterisk will be published in 1978 in the Monograph: Geology and Geochemistry of Manganese by the Publishing House of the Hungarian Academy of Sciences in co-operation with the Schweizerbart'sche Verlagsbuchhandlung Stuttgart, FRG. These papers were abstracted in 25th IGC Abstracts, Volume 3, Sydney, 1976.

Africa: one country with 1 contribution; Australia and New Zealand with 4 contributions.

At present, scientists of 20 countries expressed their willingness and intention to collaborate within the Project joining to the different panels and subdivisions, respectively. The distribution of countries among continents is as follows: 12 countries from Europe, 3 from America, 2 from Asia, 1 from Africa as well as Australia and New Zealand, although according to the Geological Correlation Number 5, and the personal information from the IGCP Secretariat merely 13 IGCP National Committees announced so far officially their participation in the Project No. 111.

2. It may be supposed that the participation in the Project will promote the communication among scientists within the single regions, countries, too. It can rely on a greater group of scientists to collaborate in the Soviet Union, India, the USA, Brazil, Australia, New Zealand, Japan, the Federal Republic of Germany, France as it may be predicted from the announcement of scientists of different countries concerning their participation.

3. As to the role of the Project, it may be considered as an accelerator and catalysator, so to say, regarding the manganese research all over the world.

4. An interest in the Project should be shared not only by countries with manganese deposits of economic importance, having at the same time research possibilities of high level and a well trained staff of experts, but developing countries, for whom the fact of being included in the organized international cooperation can be a considerable help in exploration of manganese deposits, production, instrumental investigations, training specialists etc. But the Project calls as well specialists from countries which although have no important manganese deposits but can considerably contribute to manysided investigation with modern instrumental methods and evaluating the data and undertaking the organisation of courses, seminars keeping in line with the interests of developing countries.

5. The latest results in different aspects of manganese researches are comprised in the Monograph. It is hoped that it will be of worth for transfering the knowledge to scientists in developing countries and on the other hand the Project Staff will try with all means to initiate the developing countries, which have manganese ore deposits, to share the common work in a greater number than at present. Furthermore, it is planned to look for the possibilities of organizing seminars, courses for the experts of developing countries.

6. In the Monograph a comprehensive paper will be published on the essential data of the most important manganese oxide-hydroxide minerals. The work is to be continued in order to work out international standards for manganese ore minerals and ore textures.

7. The final and detailed programme denoting the subject areas and the way of collaboration between scientists of different countries participating in the common work will be discussed and approved in August 1978 at the IAGOD Commission on Manganese-IGCP Project No. 111 Meeting to be held in Snowbird, Alta, Utah, USA, during the Fifth Symposium of the IAGOD.

8. The Project can be a link with the NOAA Project, the CCOP/SOPAC Manganese Programme as well as with the US Inter-University Manganese Nodule Project, the IUGS Commission on Marine Geology, and last but not least the IAGOD Commission on Manganese who launched this Project.

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PROGRESS REPORT ON THE RESEARCH OF THE MANGANESE ORE DEPOSITS OF ROMANIA (1973—1976)

RADU DIMITRESCU

In the last four years, mineralogical investigations were continued for the Romanian metamorphic manganese ores. A thesis already mentioned in a previous progress report [DIMITRESCU, 1971] was printed [POPA, 1971]. A second thesis [BĂLAN, 1976] was published this year. For the Iacobeni ore deposit, it describes 70 minerals, 18 of which are for the first time mentioned for the Romanian territory and 25 of which are new for Iacobeni. The most important manganese minerals are: brostenite, manganite, nsutite, cryptomelane, pyrolusite, birnessite, wad, jacobsite, pyrophanite (oxides); rhodochrosite, ponite, Ca-rhodochrosite, Mg-rhodochrosite, oligonite, kutnahorite, Mn-calcite (carbonates); tephroite, sonolite, alleghanite, spessartine, rhodonite, pyroxmangite, Mn-aegirine, Mn-aegirine-augite, dannemorite, Fe-Mn-anthophyllite, Mg-Mn-riebeckite, Mn-stilpnomelane, manganophyllite, baementite, neotocite (silicates); alabandite (sulfide); hübnerite (wolframate). Physical constants and chemical composition were determined for most of these minerals.

The parageneses indicate 10 phases of mineral formation. Transformations underwent by the protores in the oxidation zone are also discussed.

The chemical composition of some manganese minerals was determined for other ore deposits, too (Delineşti, Răzoare=Macskamezö, Baia de Arieş=Offenbanya), by POPA, COCÎRŢĂ and DIMITRESCU. They include rhodochrosite, rhodonite, pyroxmangite, bustamite, dannemorite, spessartine and alabandite.

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